Arsenic Contamination Status in Europe, Australia, and Other Parts of the World

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Abstract This chapter presents latest research in Australia, Europe, and other parts of the world on environmental issues related to As, particularly in water, but the viewpoints of food, health, and soil professionals are presented too. Having summarized more than 150 ecogeochemistry papers, the text is showcasing developments in this fast-moving field of research witnessing inadvertent arsenic poisoning on mass scale. In Europe, there are several regional hotspots of As contamination which warrant further detailed investigations. Most notable is the case of the Pannonian Basin (Hungary, Serbia, and Romania), where more than 600,000 residents are at risk of drinking water containing high As concentrations. Other regions threatened by waterborne As include Czech Republic, Croatia, Finland, Greece, Italy, Spain, and Turkey. While the majority of problems associated with arsenic mobilization in Asian regions are linked to natural processes, those recorded in Australia and New Zealand arise from both the natural processes and anthropogenic activities related to the mining industry, waste disposal, usage of arsenic pesticides and herbicides, atmospheric deposition, and timber treatment practices. Not only have the mining of mostly gold deposits and the associated gold extraction activities increased the release of As into the environment, but they also left a long-lasting legacy of the As-contaminated environment. In Africa, elevated As levels are found only sporadically across the continent, more as a result of the lack of research than a real absence of the problem. Although elevated arsenic concentrations have been reported in both the surface and groundwater of Africa, high As levels in surface waters generally are linked to mining operations, as well as to agricultural drains, local sediments, and disposal and incineration of municipal and industrial waste. Conversely, in groundwater, As occurrence is generally related to local geology, mineralization, geothermal water, etc. In Russia, drinking water quality is, in general, rather low due to surface contamination; lack of sanitary protection; delayed repair, cleaning, and disinfection of wells; and interruptions. The

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occurrences of high arsenic in soil and drinking water, although based on small number of studies, are associated with both the geogenic and anthropogenic sources. Similar situation is also found in many countries of Eastern, Central, and Western Asia where As contamination is evidenced, although only sporadically, in both drinking water and food.

Keywords Arsenic · Drinking water · Food · Contamination · Europe · Australia · Africa · Russia

1 Introduction

Arsenic is a toxic, non-essential element, used as a pigment, a livestock growth promoter, and as a component in pesticides and wood preservatives. It has caused considerable environmental pollution due to the frequent and long-term use of its compounds in industry and agriculture. It is ubiquitous in nature and is found in detectable levels in nearly all environmental media (Reimann et al. 2009). It is concentrated in volcano-sedimentary ore deposits in various mineralizations. Its freshwater content is typically 2 μg/L. Arsenic is mobile in low pH and reductive conditions. In humans, due to its chemical similarity with P, it can disrupt metabolic pathways involving P. Since adults drink on average 1.5 L/day drinking water, they are more likely affected by pollutants from this route than from the diet. Drinking water laden with As is slowly poisoning humans, starting with keratoses and discoloration of the skin, while later these turn into cancers, and liver and kidneys deteriorate.

The reasons why aquifers are contaminated by As are manifold. Mostly, it is associated with iron-rich sediments from the rivers like Ganges, Brahmaputra, Meghna, etc. Also, As is included in sulphide minerals; e.g. pyrite is the most widespread source of As. Reducing conditions lead to solubilization of Fe and the concomitant release of the adsorbed arsenate. The problem of the release of As from sediments into the groundwater is found in many parts of the world, which is elaborated in this chapter.

Strong regulatory safe drinking water standards have prompted scientists and technologists in search for water treatment schemes to remove As and other contaminants from wells used by communities in affected regions (Katsoyiannis et al. 2014; Sharma et al. 2017). Furthermore, some scientific groups employ redox active molecules against As toxicity in rice, in order to cut down the As load in rice grains and enhance their tolerance capacity along with nutritional quality (Upadhyay et al. 2018).

Environmental contamination with arsenic on global, regional, and local levels has been increasingly recognized by scientists and public authorities across the

Table 1 Arsenic maximum concentrations in different sampling water media in European (EU) regions/countries

Region	Country	µg As/L	Sample media	References
Balkan	Croatia	611	Drinking water	Ćavar et al. (2005)
	Serbia	8000	Mineral and thermal water	Dangić (2007)
		420	Public water supply	Jovanović et al. (2011)
	Greece	1840	Public water supply	Kouras et al. (2007)
	Romania	175	Drinking water	Neamtiu et al. (2015)
	Turkey	911	Groundwater	Gemici et al. (2008)
		594	Geothermal water	Gunduz et al. (2010)
East EU	Czech Republic	1141	Groundwater	Drahota et al. (2009)
	Slovakia	90,000	Sb-As-Au mine borehole	Flakova et al. (2012)
		2150	Mine surface water	Hiller et al. (2012)
	Hungary	28.6	Drinking water	Hough et al. (2010)
		88.0	Drinking water	Lindberg et al. (2006)
North EU	Sweden	17.0	Lakes	Jacks et al. (2013)
		14.0	Unimpacted surface water	Routh et al. (2007)
	Finland	2230	Drilled bedrock well	Parviainen et al. (2015)
South EU	Spain	890	Spoil heap surface water	Alvarez et al. (2006)
		118	Drinking water	Medrano et al. (2010)
	Portugal	7.8	Well water	Candeias et al. (2014)
		24.3	Surface water	Luís et al. (2011)
West EU	UK	21.0	Surface water	Camm et al. (2004)
	Ireland	234	Drinking water	McGrory et al. (2016)
	Denmark	25.3	Drinking water	Monrad et al. (2017)

world recently. Namely, arsenic contamination of underground water reservoirs has gone unrecognized for decades. Arsenic contamination of groundwater and plants in India is well documented (Srivastava et al. 2016). According to some estimates, only in West Bengal the population of 6×10^6 has been exposed to unacceptably high levels of As in drinking water (Nordstrom 2002), while many times more in Bangladesh. Since there are countless reservoirs plagued by high As levels across the world, mitigation plans require massive efforts and knowledge of a current arsenic status in the environment. The aim of this chapter is to present latest research in Europe (selected papers summarized in Table 1), Australia, and other parts of the world (selected papers shown in Table 2) on environmental issues related to As, particularly in water, but the viewpoints of food and health professionals are presented too. Through a summary of more than 150 ecogeochemistry papers, the text is showcasing developments in this fast-moving field of research witnessing inadvertent arsenic poisoning on mass scale.

Table 2 Arsenic maximum concentrations in different sampling water media in Australia, New Zealand, Africa, Russian Federation, and selected Asian countries

Region	Country/state	µg As/L	Sample media	Reference
Australia and New Zealand	New South Wales	30,000	Groundwater	Binning et al. (2001)
		300	Groundwater	Smith and Jankowski (2001)
		70	Groundwater	McLean and Jankowski (2001)
	Victoria	2830	Surface water	DMID (1991)
		5000	Surface and groundwater	Hinwood et al. (1999)
		73	Drinking water	Hinwood et al. (2003)
	Western	220,000	Groundwater	Throssell and Blessing (2001)
	Australia	7300	Groundwater	Appleyard et al. (2006)
		800	Domestic well water	
	New Zealand	47.5	Groundwater	Environment Waikato Technical Report (2006)
			Geothermal water	
Africa	Botswana	116	Groundwater	Huntsman-Mapila et al. (2006)
		3200	Groundwater	Huntsman-Mapila et al. (2011)
		180	Groundwater	Mladenov et al. (2013)
	Burkina Faso	1630	Groundwater	Smedley (1996)
	Ethiopia	96	Surface and groundwater	Reimann et al. (2003)
		566	Surface water	Rango et al. (2013)
		190	Groundwater	
	Ghana	175	Surface water	Smedley (1996)
		64	Groundwater	
		8250	Surface water	Serfor-Armah et al. (2006)
		73	Surface water	Asante et al. (2007)
		4	Groundwater	
		1760	Groundwater	Kusimi and Kusimi (2012)
	Nigeria	6.88	Groundwater	Asubiojo et al. (1997)
		28	Tap water	
	South Africa	12.3	Groundwater	Dzoma et al. (2010)
	Togo	6460	Surface water	Rezaie-Boroon et al. (2011)
Eurasia	Russian Federation	9300	Groundwater	Ilgen et al. (2011)
			Geothermal water	
		230	Groundwater	Kurbanova et al. (2013)
		59	Drinking water from well	Yurkevich et al. (2017)

(continued)

Table 2 (continued)

Region	Country/state	µg As/L	Sample media	Reference
		89	Drinking water from water-pump	Bortnikova et al. (2018)
Central Asia	Iran	1500	Drinking water	Barati et al. (2010)
		689	Groundwater	Keshavarzi et al. (2011)
		310	Drinking water	Keshavarzi et al. (2015)
		1440	Surface and ground water	Mosaferi et al. (2017)
Far East Asia	Japan	293	Well water	Kondo et al. (1999)
		17	River water	Miyashita et al. (2009)
		750	Hot spring water	
	South Korea	167	Surface and groundwater	Woo and Choi (2001)
		1.5	River water	Hong et al. (2018)

2 Europe

2.1 Balkan Countries

2.1.1 Croatia

Inorganic arsenic (iAs) in groundwater exceeding 10 µg/L (maximum contaminant level, MCL) set by the World Health Organization (WHO) is characteristic for intermediate-depth aquifers over large areas of the Pannonian Basin in Central Europe. Eastern part of Croatia belongs to the Pannonian Basin, where communities are exposed to toxic levels of As in drinking water. Groundwater, the main source of drinking water for the population in this area, contains high iron, manganese, ammonia, organic substances, and arsenic levels. Its inorganic As comes mainly from natural geological sources such as alluvial basins containing deep sediments from the Middle and Upper Pleistocene. Almost 200,000 people in eastern Croatia are daily drinking water with As concentrations ranging 10–610 µg/L. Therefore, Romić et al. (2011) sampled groundwater from 18 water wells and 12 piezometers with depth ranges 21–200 m. During the 10-year period to 2007, mean As concentration of 240 µg/L was found. Higher mean As levels (205 µg/L) were found in deeper wells (>50 m) compared to 27 µg/L in shallow groundwater (<50 m). Significant seasonal variations of total As were observed in some wells, with the highest concentrations often found during the summer. Similarly, Ujević Bošnjak et al. (2012) collected groundwater samples from 56 production wells in 2 eastern Croatian counties, Osijek-Baranja and Vukovar-Srijem. Arsenic was detected in 46 out of 56 groundwater samples, having total levels up to 491 µg/L. Also, 36 wells were found to have total As concentrations above the WHO Maximum Contaminant Level for As in drinking water, i.e. 10 µg/L. Only inorganic As species were detected, with arsenite As(III) as the predominant form. The authors interpreted As mobilization in terms of reductive dissolution of Fe oxides, desorption of As from Fe oxides and/or clay minerals, as well as competition for the sorption sites with organic matter and phosphate. Moreover, two medical studies were conducted in the same region in order to reveal some potential health issues caused by As. Ćavar et al. (2005) found the positive correlation between As in drinking water and residents hair. The mean As values in community drinking water samples were 0.14, 37.88, 171.60, and 611.89 μg/L, while the corresponding mean levels of As in hair were 0.07, 0.26, 1.74, and 4.31 μg/g. Bošnjak et al. (2008) examined prevalence and serum levels of selected markers of cardiovascular disease in 34 subjects from a rural population exposed to high drinking water As levels (611.89 ± 10.06 μg/L). The results suggest that chronic high As intake is associated with higher prevalence of obesity, hypertension, and several markers of cardiovascular disease in the exposed population.

Due to toxicity and mutagenicity, arsenic is regarded a principal pollutant in water used for drinking. Therefore, Radić et al. (2016) conducted toxicological and chemical evaluation of groundwater samples from eastern Croatia, obtained from drinking water wells enriched in As prior and following the electrochemical and ozone-UV-H2O2-based advanced oxidation processes (EAOP). They employed acute toxicity test with *Daphnia magna*, and chronic toxicity test with *Lemna minor* L., along in vitro bioassays using human peripheral blood lymphocytes (HPBLs). Several oxidative stress parameters were estimated in *L. mino*r. Untreated groundwater caused only slight toxicity to HPBLs and *D. magna* in acute experiments. However, 7-day exposure of *L. minor* to raw groundwater induced genotoxicity, a significant growth inhibition, and oxidative stress injury. EAOP treatment was highly efficient in the removal of As, ammonia, and organic compound and the complete elimination of the observed genotoxicity and toxicity of raw groundwater.

Regarding arsenic in food, Sapunar-Postružnik et al. (1996) determined total As levels in food items sold in Croatia during a 5-year period (1988–1993). Highest As mean levels were recorded in the following food groups: fish (498 μg/kg), fishery products (270 μg/kg), and cheese and dairy products (39 μg/kg). Lowest As levels were found for fruit, chocolate, and fruit products (0.2, 0.2, and 0.3 μg/kg, respectively). The authors estimated that a mean weekly dietary As intake was 81.9 μg/individual, significantly lower than in other countries, due to lower consumption of fish and fishery products in Croatia.

2.1.2 The FYR of Macedonia

The FYR of Macedonia is located in the Serbo-Macedonian Massif of the Eurasian Tethys metallogenic belt. The region contains some disused As and Sb mines (Lojane, Alchar, and Krstov Dol) which were studied by Alderton et al. (2014). They found out that soil and river sediments contained up to several hundred mg/kg of both elements, while local alkaline water contained up to hundreds of μg/L of the both. They showed how alkaline water contributes to the breakdown of As- and

Sb-sulphides and element mobility over larger areas. The Alchar locality is known as As-Sb-Tl-Au volcanogenic hydrothermal deposit, situated at the north-western margins of Kožuf mountain, close to the border between the FYR of Macedonia and Greece. There, Stevanović et al. (2010) investigated the uptake and distribution of arsenic in three *Viola* endemic species. Total As content in soil ranged from 3347 to 14,467 mg/kg, while plant available As ranged from 23 to 1589 mg/kg. The As concentration of roots ranged from 783 mg/kg in *Viola macedonica* to 2124 mg/kg in *Viola arsenica*. Only a small amount of As (<100 mg/kg) was accumulated in the aboveground parts of these species. Also, Kostik et al. (2014) determined As concentrations up to 26.4 μg/L in drinking water coming from the Kožuf mountain, as a consequence of minerals like lorandite, orpiment, realgar, arsenopyrite, etc.

2.1.3 Serbia

Central parts of the Balkan Peninsula in south-east Europe, in Serbia, and in Bosnia, mostly in the Danube River Basin, include several natural and anthropogenic sources of As, such as geological formations, ore deposits, old and active mines, and various industries. The area is also rich in mineral and thermal waters (MTWs). According to Dangić (2007), MTWs are famous for being a rich source of Fe-As medicinal water (Crni Guber in Srebrenica). Arsenic concentrations in 153 MTWs varied from <0.01 to 8 mg/L, but levels up to 30 μg/L predominate. The study showed the correlations among As levels, mineralization, Fe content, and pH in MTWs.

The first study on the distribution of As in public water supply systems in Vojvodina (the northern region of Serbia, a part of the Pannonian Basin), where aquifers contain high As values, was carried out by Jovanović et al. (2011). Around 63% of all collected waters exceeded Serbian and European standards for As in drinking water. Levels of As were much higher than in other Pannonian Basin countries, being similar to some values in Bangladesh. Large variations in As levels among water supply systems were observed (median ranged from 2 to 250 μg/L, while max values ranged from 5 to 349 μg/L). About 50.4% of them had As concentrations 11–50 μg/L, and further 38.6% systems had mean arsenic levels over 50 μg/L. The differences among the investigated municipalities were statistically significant ($p < 0.001$). Škrbić et al. (2013) examined As, Cd, and Pb in 114 samples of mostly consumed foodstuffs collected at supermarkets in Novi Sad, the capital of Vojvodina. The highest concentrations were found for Pb in candy (0.323 mg/kg), Cd in cayenne (0.118 mg/kg), and As in canned fish (0.43 mg/kg). The highest intake was estimated for Pb, 72.3 μg per day for adult population, while intakes of As and Cd were significantly lower (21.9 μg per day and 11.5 μg per day, respectively). The levels of As, Cd, and Pb in investigated foodstuffs were in compliance with the current Serbian and EC legislations.

Pontic shad (*Alosa immaculata* Bennet 1835) is an anadromous species that lives in the heavily polluted north-western part of the Black Sea and migrates into the Danube River to spawn. A total of 48 samples of Pontic shad were collected by

Višnjić-Jeftić et al. (2010) at a 863th km of the Danube River (downstream of the Djerdap II dam) to analyse content of Al, As, Cd, Cu, B, Ba, Fe, Mg, Sr, Zn, Li, Co, Cr, Mn, Mo, Ni, and Pb. Muscle samples had the lowest concentrations of most of the analysed elements, but they were characterized by the highest As levels. The study showed that As and Cd in Pontic shad muscle tissue were above maximum acceptable concentrations for human consumption, and precautions were necessary.

2.1.4 Greece

Gamaletsos et al. (2016) reviewed the existing data about the geological sources of As in Greece which include As-containing ores in active and abandoned mining areas, geothermal/hydrothermal waters, lignites in exploited and unexploited deposits, As-minerals in various rock types such as metamorphic rocks, and mineral dust originating from Sahara desert. It is considered that As release from the above sources, in conjunction with various anthropogenic As fluxes, occasionally creates distinct areas with contaminated groundwater, soils, marine, and atmospheric environment. They emphasize that the most important and persisting sources of As exposure to the Greek population are geothermal and hydrothermal fluids arising from faults as well as the volcanic activity. Eliopoulos et al. (2012) studied iron-nickel laterite deposits in the Balkan Peninsula and Turkey, located in the Mirdita-Sub-Pelagonian and Pelagonian geotectonic zones, extending into the Anatolides zone. Their arsenic contents are generally low, ranging from less than 2 to a few tens of mg/kg. However, in the Aghios Ioannis deposit (Lokris, Central Greece), As attains values up to 0.26 wt.% As. In Western Turkey, As ranges from 0.004 to 1.07 wt.% in the Gordes deposit. The authors emphasize that elevated As contents in goethite (Fe oxides) in Fe-Ni laterites of Greece and Turkey, due to its absorption capacity, are considered to be of particular significance in the remediation of aquifer and soil contamination rather than being a source of environmental risk.

Regarding groundwater, Katsoyiannis et al. (2014) summarized the arsenic contamination in Europe, where several countries are affected by elevated arsenic concentrations (i.e. Greece, Hungary, Romania, Croatia, Serbia, Turkey, and Spain). In Greece, arsenic-affected regions are classified in three categories: (1) the geothermal-affected waters, (2) the alluvial deposits of rivers and aquifers, and (3) those influenced by mineralization/mining activities. In Greece, As levels in geothermal waters vary from 30 to 4500 μg/L, in the regions close to alluvial deposits from 15 to 100 μg/L, and in areas affected by mining activities from 20 to 60 μg/L. Katsoyiannis and Katsoyiannis (2006) investigated groundwater used for drinking water supply in the greater industrial area of Thessaloniki (Northern Greece), where As levels exceeded the WHO provisional guideline value and the EU maximum contaminant level of 10 μg/L. The concentration of total arsenic was in the range 4–130 μg/L (median was 36 μg/L), while 9 out of 11 wells contained total As >10 μg/L. The examined groundwater contained elevated concentrations of manganese and phosphate. Furthermore, Kouras et al. (2007) analysed groundwater in the area of

Chalkidiki (Northern Greece) where As showed high spatial variation with range from 0.001 to 1.840 mg/L. Almost 65% of the examined groundwater samples had As >10 μg/L. Arsenic was highly correlated with potassium, boron, bicarbonate, sodium, manganese, and iron, suggesting common geogenic origin of these elements and conditions that enhance their mobility. Aloupi et al. (2009) studied the occurrence of As in water from a drainage basin of Kalloni Gulf (island of Lesvos, Greece). The aim of the study was to investigate the potential influence of the geothermal field of Polichnitos-Lisvori on the ground and surface water systems of the area. Total dissolved As varied in the range <0.7–88.3 μg/L in groundwater, 41.1–90.7 μg/L in thermal spring water, and 0.4–13.2 μg/L in stream water. Four out of 31 groundwater samples exceeded the EC standard of 10 μg/L.

Casentini et al. (2011) investigated the accumulation of As in soils and food crops due to the use of arsenic-contaminated groundwater for irrigation in the Chalkidiki prefecture. The groundwater As levels are >1000 μg/L within the Nea Triglia geothermal area. Arsenic content in sampled soils ranged from 20–513 to 5–66 mg/kg inside and outside of the geothermal area, respectively. Low As accumulation was found in collected olives (0.3–25 μg/kg in flesh and 0.3–5.6 μg/kg in pits). The authors pointed out that arsenic uptake in fast-growing plants should have been assessed as soil As values were frequently elevated far above recommended levels.

2.1.5 Cyprus

During 2007–2009, there was a national monitoring programme in Cyprus, when 84 boreholes were sampled and analysed for total arsenic by ICP-MS (Christodoulidou et al. 2012). The groundwater As concentrations ranged from <0.3 to 41 μg/L. Several boreholes were repeatedly sampled following an initial borehole washout for 5 min. Further sampling in 2010 revealed that total As concentrations ranged <0.3–64.2 μg/L, with arsenate (As^V) as the predominant As species (determined by a novel field-based methodology). The study showed that maximum total As concentration was sixfold the WHO drinking water guideline limit (10 μg/L) and approximately half the United Nations Food and Agriculture Organization irrigational limit of 100 μg/L As.

2.1.6 Romania

In Western Romania, near the border with Hungary, the Arad, Bihor, and Timis counties use drinking water coming partially or entirely from iAs-contaminated aquifers. More than 45,000 people residing in the Arad and Bihor counties are exposed to iAs above 10 μg/L via public drinking water sources. Neamtiu et al. (2015) analysed iAs in 124 public and private Timis County drinking water sources, including wells and taps, used by pregnant women participating in a case-control study of spontaneous loss. Arsenic levels in wells (median 3.1 μg/L, range

<0.5–175 μg/L) were higher than As in community taps (median 2.7 μg/L, range <0.5–36.4 μg/L). The results showed that pregnant women from the Timis County, compared to surrounding counties, were exposed to lower arsenic levels via drinking water. Butts et al. (2015) conducted a pilot study of associations between iAs in drinking water and self-reported chronic diseases in 297 pregnant women from the Timis County. Adjusted for confounding variables, they identified the positive association among iAs and heart disease, kidney disease, and high blood pressure. Also, Bloom et al. (2016) examined birth outcomes in the Timis County where they followed 122 women with singleton deliveries, for whom they constructed individual exposure indicators using self-reported water consumption weighted by arsenic measured in drinking water sources. They found out no overall confounder-adjusted effects for arsenic exposure on birth outcomes. Yet, their results suggest smoking may potentiate an otherwise benign arsenic exposure.

In the same Timis County, Susko et al. (2017) evaluated associations among low As levels in drinking water and female fecundity. Women ($n = 94$) with planned pregnancies of 5–20 weeks gestation completed a comprehensive physician-administered study questionnaire and reported the number of menstrual cycles attempting to conceive as the time to pregnancy (TTP). There was no main effect for the drinking water As exposure, yet the conditional probability for pregnancy was modestly lower among arsenic exposed women with longer TTPs, relative to women with shorter TTPs, and relative to unexposed women. Although preliminary, the results suggest that low-level As contamination in residential drinking water sources may further impair fecundity among women with longer waiting times.

2.1.7 Turkey

Turkey is characterized by complex geology with active tectonics and high geothermal potential. Particularly, its western part is a region of abundant geothermal activity, which is controlled by faults accommodating the deep circulation of hydrothermal fluids of meteoric origin. Western Anatolian young (down to about 1 my) volcanic activities and shallow intrusive emplacements resulted in the formation of considerable number of geothermal and hydrothermal systems and kept them active for long enough that gave rise not only to the formation of numerous epithermal mineralizations rich in As-Sb-Hg-bearing sulphide minerals but also to the formation of As-enriched sedimentary and magmatic rocks. Their dissolution and redistribution within the rocks and resultant soils consequently made them geogenically As-rich. According to Baba and Sözbilir (2012), the highest concentrations of naturally occurring aqueous As are found in major graben faults. Herewith, elevated As levels in geothermal resources have been detected in Western Turkey, including but not limited to Biga Peninsula, Gediz Graben, Kucuk, and Buyuk Menderes Graben, with As values ranging from 1 to 1419 μg/L. Another study related to geothermal fluids was conducted by Gunduz et al. (2010). They sampled 27 wells in the surficial aquifer and found average As concentration of 99.1 μg/L, with max value of

561.5 μg/L. There, Cu-Pb-Zn mine was in operation in the past, and arsenic-containing waste (660 mg/kg As) was deposited in an uncontrolled manner. Another potential source of As in the study area is geothermal fluid reaching As levels as high as 594 μg/L. Consequently, death statistics from the 1995 to 2005 period collected from the study area revealed increased rates of gastrointestinal cancers above Turkish average.

Ozkul et al. (2015) investigated As concentrations of soils and stream waters from an area where the world class borate deposits (Bigadiç district) were mined for about 60 years. Arsenic levels in soil and water samples ranged from 0.4 to 2488.4 mg/kg and 8–243 μg/L, respectively. Also, Gemici et al. (2008) documented the environmental impact of the borate mines, which are the largest colemanite and ulexite deposits in the world. High groundwater sulphate concentrations (up to 519 mg/L) derive from anhydrite, gypsum, and celestite minerals in the borate zone. Arsenic range (33–911 μg/L) exceeds the tolerance limit of 10 μg/L, along the suphate, Al, and Fe levels which are above the drinking water standard for groundwater samples.

Regarding medical research, Gunduz et al. (2015) compared the causes of death in five villages situated in Simav Plain, during the period 2005–2010, where groundwater was characterized by As ranges 7.1–833.9 μg/L. They formulated a two-phase research; in the first phase, public health surveys were conducted with 1003 villagers to determine the distribution of diseases, and in the second phase, verbal autopsy surveys and official death records were used to investigate the causes of death. In total, 402 death cases were found in the study area where cardiovascular system diseases (44%) and cancers (15.2%) were major causes. Cancers of lung (44.3%), prostate (9.8%), colon (9.8%), and stomach (8.2%) were comparably higher in villages with high arsenic levels in drinking water supplies. Furthermore, the majority of cases of liver, bladder, and stomach cancers were observed in villages with high arsenic levels.

2.2 Eastern Europe

2.2.1 Poland

In Poland, soil-arsenic problem is mainly confined to industrial, mining, and smelting areas. Karczewska et al. (2007) indicated a few “hot spots”, where soils are characterized by extremely high levels of As, caused by natural enrichment and long-lasting ore mining and processing activities. They presented total concentrations and speciation of As in Zloty Stok, Zelezniak, and Czarnow soils (SW Poland), where As ores were mined and processed since the thirteenth century. Maximum concentrations of As in mine spoils at the Zloty Stok, Czarnow, and Zelezniak sites were 8800, 40,600 and 18,100 mg/kg, respectively. Alluvial soils in these three regions contained up to 11,500, 4210 and 6800 mg/kg of As, respectively.

As regards the Zloty Stok mine, it is located in the southern part of a town of the same name in Lower Silesia. The exploitation of arsenic started in the eighteenth century. Arsenic occurs as loellingite (FeAs2) and arsenopyrite (FeAsS) in serpentinite and marbles. Drewniak et al. (2008) investigated whether bacteria present in ancient gold mine could transform immobilized arsenic into its mobile form and increase its dissemination in the environment. Twenty-two arsenic-hypertolerant cultivable bacterial strains were isolated. A correlation between the presence of siderophores and high resistance to arsenic was found. The results of the study showed that the detoxification processes based on arsenate reductase activity might substantially disseminate the arsenic pollution. The authors concluded that the activity of the described heterotrophic bacteria contributed to the mobilization of As in the more toxic As(III) form, and this had led to the contamination of important drinking water sources situated around the mine and mine waste deposits. There, Jedynak et al. (2012) assessed the strategy developed by terrestrial plants to avoid or minimize the toxic effects caused by As. They found out that the highest concentration of arsenic was found in herb Robert (*Geranium robertianum*) 21 mg/kg and common nettle (*Urtica dioica*) 5.3 mg/kg in the cases of Złoty Stok and Łomianki, respectively. Phytochelatins were present in all investigated plant species: PC3 was present in the highest concentration in plants from Złoty Stok (compared to other phytochelatins), while none of the phytochelatins dominated in plants from Łomianki.

In addition to natural sources and land-originated pollution, the Baltic Sea has another anthropogenic source of arsenic in bottom sediments – arsenic-based chemical warfare agents (CWA). Beldowski et al. (2015) reported elevated As levels in dumpsite areas compared to reference areas. However, very complex and variable chemical transformations of arsenic and As-containing compounds in the marine environment complicate the interpretation of sedimentary concentration data for arsenic and its compounds and hinder the establishment of any association with chemical weapons.

In terms of food research, Niedzielski et al. (2013) determined the content of As(III), As(V), and DMAA (dimethylarsinic acid) in *Xerocomus badius* fruiting bodies collected from selected Polish forests from areas subjected to very low or high anthropopressure, and some commercially available samples obtained from the Polish Sanitary Inspectorate. The analyses showed high levels (up to 27.1, 40.5, and 88.3 mg/kg for As(III), As(V), and DMAA, respectively) of arsenic, and occurrence of different species in mushrooms collected from areas subjected to high anthropopressure and two commercially available samples. Therefore, the authors suggested the need for monitoring of As in mushroom foodstuffs.

2.2.2 Czech Republic

In Central Bohemia, approximately 50 km south of Prague, there the Mokrsko-West gold deposit is situated, never mined though (Bohemian Massif). It is characterized by a low sulphide content (generally <3 vol.%), while arsenopyrite (FeAsS) is the predominant sulphide, and its weathering flux corresponds to approximately 95% of the total input of As into the soil. High concentrations of As in the groundwater (255–1690 μg/L) and surface water (50–340 μg/L) were measured in shallow wells and a stream, respectively. Drahota et al. (2009) determined the processes that caused a release of As into the water and its speciation under various redox conditions. In soils, As was found to be associated mainly with secondary arseniosiderite, pharmacosiderite, Fe oxyhydroxides, and rarely scorodite. Under oxidizing conditions, surface waters were undersaturated with respect to arsenate minerals, which promoted dissolution of secondary arsenates and increased As concentrations up to 300–450 μg/L in the stream and fishpond waters. According to the authors, microbial processes caused the transformation of aqueous As species as well as the mobility of As.

The Kutná Hora (Central Bohemia) is an area characterized by former (mediaeval) silver mining activity. Kralova et al. (2010) determined total element values in soils, i.e. 30 mg/kg As, 1.0 mg/kg Cd, 200 mg/kg Cr, 80 mg/kg Ni, 140 mg/kg Pb, and 200 mg/kg Zn, which exceeded the Czech threshold values by up to 15-, 30-, and 80-fold for zinc, cadmium, and arsenic, respectively. Diversity of vegetation cover (29 species) growing at the contaminated soil close to the former mine was ordinary, without occurrence of metallophytes. The element concentrations in aboveground biomass were low. Transfer factors (ratio of element content in plant and its "pseudo-total" content in soil) varied from 0.0003 to 0.003 for As, 0.001 to 0.174 for Cd, and 0.016 to 0.169 for Zn.

Majzlan et al. (2014) reported findings on extremely As-rich acid mine waters caused by weathering of native arsenic in the sulphide-poor environment of the Svornost mine in Jáchymov. Arsenic was rapidly oxidized to arsenolite, and droplets of liquid on the arsenolite crust had high As concentration (80,000–130,000 mg/L) and pH close to 0. The authors were able to isolate microorganisms on oligotrophic media with pH ~1.5 supplemented with up to 30 mM As(III). These microorganisms were adapted to highly oligotrophic conditions which disabled long-term culturing under laboratory conditions. The extreme conditions make this environment unfavourable for intensive microbial colonization, but the first results of this study showed that certain microorganisms could adapt even to such harsh conditions.

2.2.3 Slovak Republic

In the eastern part of the Slovak Republic, there is a significant source of contamination with As originating from the deposited coal fly ash, located near the village of Poša. The As pollution of the area has been reported since 1995, representing one of the most serious environmental risks in Slovakia (Hiller et al. 2009). There, the water of Kyjov Brook was characterized by a high content of total As ranging from >300 to 11,000 μg/L. Combined results of the column leaching, batch extraction, sequential extraction tests, and mineralogical analysis showed that As mobilization potential from sediments was likely controlled by Fe, Al, and Mn oxides and by pH. Plants overlying the impoundment had As concentrations 10–100 times above those from reference sites.

A similar study was conducted by Jurkovič et al. (2011) at the village of Zemianske Kostolany and 4 km south of a thermal power plant at Novaky, Prievidza district in central Slovakia. Soils at the study area were severely contaminated with arsenic after dam failure of the coal-ash pond. Mean As concentrations of soil samples collected from three sampling depths (0–20, 20–40, and >40 cm) were 173, 155, and 426 μg/g, respectively, exceeding greatly the Dutch intervention threshold.

Flakova et al. (2012) studied arsenic and antimony contamination at the Pezinok mining site (SW Slovak Republic). The highest dissolved As concentrations correspond to mine tailings (up to 90,000 μg/L), and arsenic was present predominately as As(V). Arsenic and antimony are transported by groundwater flow towards the Blatina Creek, where arsenic and antimony are attenuated by dilution and adsorption on ferric iron minerals in stream sediments.

Environmental contamination with As and Sb, caused by past mining activities at Sb mines, is a significant problem in Slovakia. Hiller et al. (2012) investigated environmental effects of five abandoned Sb mines on water, stream sediment, and soil close to residential areas. Mine wastes had up to 5166 mg/kg As, while water contained up to 2150 μg/L As. Leachates from mine wastes contained as much as 8400 μg/L As, suggesting that mine wastes could have a great potential to contaminate the downstream environment.

Drličkova et al. (2013) carried out a study of the growth of maize (*Zea mays* L.), hybrid Valentina, from an old Sb-Au mining area near Pernek in Male Karpaty Mountains. The below- and aboveground tissue As concentration, as well as toxic effect of As, was more prominent in plants grown in heap soil containing considerably less total As than Ochre soil. According to the authors, this paradox could be possibly explained in terms of decreased mobility of As in ochre substrates which are able to adsorb arsenate to Fe-(hydr)oxides. These compounds are able to reduce As concentration in soil solution and hence its bioavailability for plants.

Similarly, Vaculik et al. (2013) investigated selected medicinal plants (*Fragaria vesca*, *Taraxacum officinale*, *Tussilago farfara*, *Plantago major*, *Veronica officinalis*, *Plantago media*, and *Primula elatior*) naturally growing on old mining sites contaminated by As and Sb. Their levels in shoot ranged between 1 and 519 mg/kg for As and 10 and 920 mg/kg for Sb. The authors determined differences in the bioaccumulation of As and Sb as well as in their translocation from root to shoot within the same species growing on different localities. They emphasized that the increased bioaccumulation of As and Sb in biomass of investigated plants might be dangerous for humans when used for traditional medicinal purposes.

In terms of health studies, Ranft et al. (2003) assessed arsenic exposure of a population living in the vicinity of a coal-burning power plant with high arsenic emission in the Prievidza District. In total, 548 spot urine samples were speciated for inorganic As (Asinorg), monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), and their sum (Assum). There was a significant but weak association between As in soil and urinary Assum ($r = 0.21$, $p < 0.01$). Persons living in the vicinity of the plant had 27% higher Assum values ($p < 0.01$).

2.2.4 Hungary

Lindberg et al. (2006) published results of the study which was the part of a large case-control study of cancer risks in relation to arsenic exposure via drinking water in Central Europe: ASHRAM – Arsenic Health Risk Assessment and Molecular Epidemiology. Study areas included a few counties in Hungary (Bacs, Békés, Csongrad, and Jasz-Nagykun-Szolnok), Romania (Bihor and Arad), and Slovakia (Banska Bystrica and Nitra) with known hotspots of arsenic in drinking water. In total, 520 individuals were investigated by measuring inorganic arsenic and methylated arsenic metabolites in urine. Significantly higher concentrations of arsenic were found in both the water and the urine samples from Hungary compared to Slovakia and Romania. Significant correlation was found between water and urine As. At low water arsenic concentrations, the relative amount of dimethylarsinic acid (DMA) in urine was increased, indicating exposure via food. High body mass index was associated with higher concentrations of arsenic in urine, mostly in the form of DMA. Smokers had significantly higher urinary arsenic concentrations than nonsmokers. According to the authors, exposure to As from food, mainly as DMA, and cigarette smoke, mainly as inorganic arsenic, are major determinants of arsenic exposure at very low concentrations of arsenic in drinking water. Furthermore, Hough et al. (2010) reported their results collected under the auspices of the ASHRAM. The proportions with average lifetime exposure over 50 μg/L varied in the three countries from 1% to 16%. The proportion of population currently exposed to >50 μg/L is now between 0% and 1%, reflecting the mitigation efforts that have been made.

In terms of food safety, Soeroes et al. (2005) carried out arsenic speciation analysis on freshwater farmed fish collected from an area with elevated groundwater As concentrations as well as outside of the area. In the catfish, the accumulated total arsenic (2510–4720 μg/kg) was found mostly in the form of arsenobetaine, confirming that an uptake of As was dominated by their diet. Carp (*Cyprinus carpio*) were cultured in surface lakes with no significant arsenic pollution, containing total As concentrations 62–363 μg/kg.

Since inorganic As can get easily through the placenta, the aim of the study conducted by Rudnai et al. (2014) was to explore the associations between As content of drinking water and prevalence of some congenital anomalies. The results showed an increased risk of congenital heart anomalies among infants whose mothers were exposed to drinking water with arsenic content above 10 μg/L during pregnancy.

2.3 Scandinavia

2.3.1 Finland

In Finland, the 3-year (2004–2007) RAMAS project gathered diverse As information and identified the risk areas from the Tampere-Häme region (SW Finland), where As levels are above Finnish average As concentrations in bedrock and soil. It also provided certain risk management guidelines, mainly from groundwater and health perspectives. The subsequent ASROCKS project (2011–2014) continued the work and focused on guidelines for the rock aggregate and construction industries. Parviainen et al. (2015) published a brief review of the main results of these projects.

Abass et al. (2017) carried out a study under the auspices of the NFBC (Northern Finland Birth Cohort) programme, in order to analyse the blood levels of arsenic (B-As), cadmium (B-Cd), lead (B-Pb), total mercury (B-Hg), and selenium (B-Se) and to correlate them with calcium and haemoglobin (249 NFBC subjects). Among them, 23% of males and 17% of females had B-As levels above the ATSDR (Agency for Toxic Substances and Disease Registry) normal human levels of B-As in unexposed individuals (1.0 μg/L). Arsenic levels were elevated, statistically significantly, among those with lower reindeer, moose, and wildfowl consumptions ($p = 0.018$).

Rintala et al. (2014) reported total and inorganic As levels in long-grain rice and rice-based baby foods on Finnish market. Inorganic arsenic levels in long-grain rice varied from 0.09 to 0.28 mg/kg, and total As levels ranged from 0.11 to 0.65 mg/kg. Total As levels of rice-based baby foods ranged from 0.02 to 0.29 mg/kg. The authors estimated that inorganic arsenic intake from long-grain rice and rice-based baby food in Finland indicates that in every age group, the intake is close to the lowest $\mathrm{BMDL}_{0.1}$ value of 0.3 μg/kg bw/day, set by the EFSA.

2.3.2 Sweden

Some 10–15 years ago, Sweden had begun remediation of its many contaminated sites, a process that would cost an estimated SEK 60,000 million (USD 9100 million). Although the risk assessment method, carried out by the Swedish EPA, was driven by health effects, it did not consider actual exposure. Instead, the sites were assessed based on divergence from guideline values. Therefore, Forslund et al. (2010) used an environmental medicine approach that took exposure into account to analyse how cancer risks on and near arsenic-contaminated sites were implicitly valued in the remediation process. The results showed that the level of ambition was high. At 23 contaminated sites, the cost per life saved varied from SEK 287 million to SEK 1,835,000 million, despite conservative calculations that in fact probably underestimated the costs. It was concluded that if environmental health risks were to be reduced, there were probably other areas where economic resources could be used more cost-effectively.

Weathering of mine tailings have resulted in high As concentrations in water (up to 2900 μg/L) and sediment (up to 900 mg/kg) samples around the Adak mine (NW Sweden). Time series-based sediment incubations were set up in the laboratory with contaminated sediments to study the microbial processes involved in transformation and remobilization of As across the sediment-water interface (Routh et al. 2007). The microcosm experiments indicated that microorganisms are capable of surviving in As-rich sediments, reducing As(V) to As(III). A decrease in total As levels in sediments is coupled with an increase in As(III) concentration in the aqueous media.

The results implied that active metabolism was necessary for As(V) reduction. According to the authors, the microorganisms possessed reduction mechanisms that were not necessarily coupled with respiration, but most likely imparted resistance to As toxicity.

Broberg et al. (2014) assessed effects of arsenic on genome-wide DNA methylation in newborns. They studied As metabolite levels in maternal urine of 127 mothers and cord blood of their infants. Urinary arsenic in early gestation was associated with cord blood DNA methylation (Kolmogorov-Smirnov test, P-value<10–15), with more pronounced effects in boys than in girls. Much weaker associations were observed with arsenic exposure in late compared with early gestation. The authors concluded that early prenatal arsenic exposure appeared to decrease DNA methylation in boys. Associations between early exposure and DNA methylation might reflected interference with de novo DNA methylation.

Jacks et al. (2013) conducted a study in northern Sweden where metasediments contain pyrite, pyrrhotite, and arsenopyrite overlain by till which has up to 100 mg/kg As, while sandy sediments may contain 100–500 mg/kg As. The cycling of As in water flora and fauna in wetlands was studied. Ferric reduction occurred in wetlands and groundwater rich in Fe, and As was found to be discharging into ditches, brooks, and streams. Wetland trees and plants showed moderately elevated As levels, and a few species had >2 mg/kg As which is the permissible level in fodder for domestic animals. The only plants with a high content of As were *Equisetum* species (up to 26 mg/kg As). Elevated As levels were found in a limited number of benthic macroinvertebrate samples (1.23–42.1 mg/kg DW). Fish species from polluted streams (pike and brown trout) had normal As levels (0.57–1.84 mg/kg DW).

Jorhem et al. (2008) carried out a survey of the levels of Cd, Pb, and As in different types of rice available on the Swedish retail market. Mean levels were the following: total As 0.20 mg/kg, inorganic As 0.11 mg/kg, Cd 0.024 mg/kg, and Pb 0.004 mg/kg. The authors concluded that in countries where rice was a staple food, it might represented a significant contribution in relation to the provisional tolerable weekly intake for Cd and inorganic As.

2.3.3 Norway

De Gieter et al. (2002) determined As levels in muscle and liver tissue of 25 sea fish and 4 shellfish species from the North Sea. Highest total As concentrations (average >20 mg/kg WW) were found in lemon sole, dogfish, ray, and witch. The same species as well as the other flatfishes contained the highest amounts of toxic As (>0.1 mg/kg WW). Toxic fractions (AsTox/AsT%) above 2% were found in the following six species: sea bass, ling, John Dory, pouting, dab, and brill. The authors pointed out that in the worst-case scenario (drying or smoking of fish when the toxic As level is high, e.g. 0.5 mg/kg WW), the As content of North Sea marine food might reach harmful levels.

Savinov et al. (2003) found highest levels of the most toxic elements Cd and Hg in birds nesting north of Spitsbergen. Extremely high levels of As were detected in tissues of all seabird species collected at colonies in Chernaya Guba (Novaya Zemlya), where nuclear tests were carried out in the 1960s. Generally, levels of all analysed trace elements in the Barents Sea seabirds were similar or lower in comparison with those reported for the same seabird species from the other Arctic areas.

Samples of complete feedingstuffs for fish, and fishmeals from the Norwegian Fish Feed Monitoring Programme in 2003 were analysed for their total arsenic and inorganic arsenic contents (Sloth et al. 2005). Levels of 3.4–8.3 and 0.010–0.061 mg/kg for complete feedingstuffs were found for total As and inorganic As, respectively. The results were in accordance with typical As levels for fish reported in the literature. The data illustrated that fish fed with high levels of total As, but low levels of inorganic As are at risk of being unnecessarily rejected from fish feed markets.

Julshamn et al. (2012) determined contents of total As and inorganic As in fillet samples of Northeast Arctic cod, herring, mackerel, Greenland halibut, tusk, saithe, and Atlantic halibut. The concentrations of total As varied greatly between fish species and ranged from 0.3 to 110 mg/kg WW, while inorganic As was very low (<0.006 mg/kg) in all cases. The authors emphasized that the obtained results questioned the assumptions made by the European Food Safety Authority (EFSA) on the inorganic As level in fish used in the EFSA opinion on arsenic in food.

The study carried out by Julshamn et al. (2013) was one of several baseline studies that would provide basic and reliable information about the contents of undesirable substances in important fish species caught in Norwegian waters. They reported levels of trace metals in muscle and liver samples of more than 800 Northeast Arctic cod caught at 32 sites in the Barents Sea. The highest total As levels were found in cod from its eastern part. Arsenic concentrations varied greatly among individual fish, ranging from 0.3 to 170 mg/kg WW in muscle. According to the authors, such high levels of total As had never been reported previously for any fish. The authors ascribed them to the shrimp in the cod's diet.

2.4 Southern Europe

2.4.1 Italy

Since the pre-Roman Age, southern Tuscany has been an important mining district in Italy. Epithermal deposits of Hg and Sb were intensely exploited until the 1970s. The intensive mining and smelting activities have resulted in huge quantities of waste which, in the absence of any reclamation, still release toxic trace elements in the local environment. Due to the fact that As is a minor but ubiquitous constituent of the epithermal mineral assemblage of those ores, arsenic also contributes to the overall pollution of the mining areas. Baroni et al. (2004) surveyed As contents of soils and higher plants at two former Sb mining areas and an old quarry once used for the ochre extraction. Total As in soil ranged from 5.3 to 2035.3 mg/kg, while soluble and extractable As ranges were 0.01–8.5 and 0.04–35.8 mg/kg, respectively. The highest As contents were found in roots and leaves of *Mentha aquatica* (540 and 216 mg/kg, respectively), and in roots of *Phragmites australis* (688 mg/kg). In general, As in plants was low, especially in crops and most common wild species.

Beni et al. (2011) investigated the effects of As-contaminated irrigation water on *Lactuca sativa* L. cropping. Two different arsenic concentrations, i.e. 25 and 85 μg/L, and two different soils, i.e. sandy and clay loam, were surveyed. They determined the As mobility in different soil fractions, its amount in groundwater, and the phytotoxicity and genotoxicity. Data indicated that at both concentrations in sandy soil, As is partly rapidly leached into the groundwater and partly absorbed by vegetables, being readily available for assimilation by consumption.

Beccaloni et al. (2013) estimated the dietary intake of arsenic, cadmium, lead, and zinc in a small town located in an industrialized area of Sardinia. Spices and herbs showed the highest element concentrations, while the highest median concentrations for other food have been found in pulses for As and Zn (0.142 and 13.03 mg/kg, respectively), in leafy vegetables for Cd (0.147 mg/kg), and in fruits for Pb (0.294 mg/kg). Human health risk assessment was evaluated for three population groups: total population, infants, and children. The authors didn't find the toxicological parameters exceeding reference values in any of the studied populations. The highest estimated intakes were found for Pb and Cd and the lowest ones for Zn and As.

Caporale et al. (2013) studied the influence of compost on the growth of bean plants irrigated with As-contaminated water and its influence on the mobility of As in the soils and the uptake of As. The bean plants exposed to As showed typical phytotoxicity symptoms, but no plants died over the study. Biomass decreased with increasing As concentrations, but the reduction in the biomass was significantly lower with the addition of compost, indicating that the As phytotoxicity was alleviated by the compost. For the same As concentration, the As content of the roots, shoots, and beans decreased with increasing compost added compared to control samples. Most of the As adsorbed by the bean plants accumulated in the roots, while a scant allocation of As occurred in the beans. Hence, the authors concluded that the addition of compost to soils could be an effective means to limit As accumulation in crops from As-contaminated waters.

Rice is comparatively efficient in terms of assimilation of inorganic arsenic (iAs) into its grain, being the dominant source of this element to mankind. Therefore, Sommella et al. (2013) investigated variation of the total arsenic (tAs) and iAs content of Italian rice grain sourced from market outlets according to geographical origin and type. Mean values of tAs concentration on a variety basis ranged from 0.18 to 0.28 mg/kg and from 0.11 to 0.28 mg/kg on the production region basis. Regarding iAs concentration, means ranged from 0.08 to 0.11 mg/kg by variety and from 0.10 to 0.06 mg/kg by region. The authors determined that there was a significant geographical variation for both tAs and iAs.

Soleo et al. (2008) characterized the different sources of exposure to arsenic. They determined urinary excretion of total As, the sum of inorganic As + MMA + DMA, As3, As5, monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), and arsenobetaine in 49 workers at a steel foundry, with presumed occupational exposure to As, and 50 subjects from the general population, all males. The results pointed at no evidence of occupational exposure to As. The urinary concentrations of As3, MMA, DMA, the sum of inorganic As + MMA + DMA, and total As were not different between the two groups, while arsenobetaine was significantly higher in control participants compared to workers. The authors concluded that their research had indicated that in populations with a high consumption of seafood characterized by a significant content of As, evidence of a considerable environmental As pollution of coastal/marine aquaculture areas as well as mineral water with a relatively high content of As, minor occupational exposure to As played a negligible role in conditioning the urinary excretion of the different species of As.

2.4.2 Spain

In the area of Riaño-Valdeburón (northern Spain), there are mineral occurrences of As, Sb, Hg, Cu, and occasionally Au and Ag. A few of them have been subjected to intermittent small-scale mining since the nineteenth century. The study carried out by Alvarez et al. (2006) was focused on soil and surface water geochemistry downstream of the Santa Águeda As mine, close to the Riaño reservoir, in the Northeast of León province. There, a water reservoir (catchments of the Esla, Yuso, and Orza Rivers) was constructed for the production of hydraulic energy and drinking water purposes. It receives leachates from polluted soils and spoil heaps from a site where small-scale As mining and smelting operations were active in the first half of the twentieth century. Although total As concentrations in soils reach 23,800 mg/kg and total As values in surface waters reach 890 μg/L, the water flow from the mine catchment has a negligible contribution when compared with the total volume of water inside the dam (0.07%).

Recently, elevated As concentrations in groundwater used for drinking water supplies have been recognized in case of the Madrid Tertiary detrital aquifer. Although natural causes have been suggested as the source of As, the study by Recio-Vazquez et al. (2011) aimed to determine whether an anthropogenic contribution could have been involved too. During the sub-catchment's areas sampling, they found out that many occurrences of arsenopyrite [FeAsS] were artificially outcropped and dumped out, and mining wastes were scattered and exposed to weathering. Several mineral and ground specimens were collected, and it was determined that their content of As was very high (113,702 mg/kg). The authors concluded that the contamination of the area due to old mining activities could release arsenic to Madrid water supplies and suggested that additional decontamination studies were necessary.

Gomez-Gonzalez et al. (2014) conducted an experiment in a shrubland situated in the upper portion of a small sub-catchment of the Guadalix River (Madrid), which feeds into the Madrid Tertiary Detrital Aquifer. They point out that the formation of scorodite leads to the natural attenuation of arsenic in a wide range of environments. However, they questioned the long-term stability of scorodite-rich waste piles. Their results indicate that the deposit behaves as an acute point source of As and confirms the strong association of As(V) with Fe(III) oxide phases. Herewith, the authors emphasize that there is a need to monitor and reclamate As-rich mine deposits.

Almela et al. (2006) determined total arsenic, inorganic arsenic, lead, and cadmium levels in 112 samples of seaweed preparations sold in Spain (seaweed packed in plastic or cardboard box, seaweed in the form of tablets and concentrates, foods containing seaweed, and canned seaweed). The concentration ranges found, expressed in mg/kg, dry weight, were total As (0.031–149), inorganic As (<0.014–117), Pb (<0.050–12.1), and Cd (<0.003–3.55). All the analysed contaminants exceeded legislated values. Particularly, all the samples of *Hizikia fusiformis* exceeded the inorganic As limit established in some countries. According to the authors, the consumption of 3 g/day of the analysed samples could represent up to 15% of the respective tolerable daily intakes (TDI) established by the WHO. They specifically emphasized that the situation was especially alarming for the intake of inorganic As from *Hizikia fusiformis*, which could be three times the TDI established.

It is well-known that there is a positive correlation between rice content and As level in foods. According to Burlo et al. (2012), this is of extraordinary importance for infants below 1 year of age as their diet is very limited, especially in case of celiac disease when affected infants consume only gluten-free products, such as rice or corn. Arsenic contents were found to be significantly higher ($P < 0.001$) in gluten-free infant rice (0.057 mg/kg) than in products with gluten (0.024 mg/kg). Moreover, the authors suggested special precaution in case of preparation of rice-based products at home as arsenic content in Spanish rice was high (>0.3 mg/kg in some cases).

A similar study was conducted by Hernández-Martínez and Navarro-Blasco (2013). They determined levels of total Hg and As in 91 various infant cereals from 10 different Spanish manufacturers. In general, the content of toxic elements (median (Q1; Q3)) found in infant cereals based on conventionally obtained raw materials ($n = 74$, Hg: 2.11 (0.42; 4.58), As: 21.0 (9.4; 50.9) μg/kg) was lower than in cereals produced by organic methods ($n = 17$, Hg: 5.48 (4.54; 7.64), As: 96.3 (87.5; 152.3) μg/kg). The highest As content was found in rice-based cereals. It was calculated that 95% of the organically produced infant cereals and 70% of the conventional gluten-free infant cereals showed an inadmissible risk of arsenic intake. The authors concluded that continued efforts in standardizing routine quality control and the reduction in As levels in infant cereals are urgently needed. Also, they call for relevant legislation to be established and regulated by EC regarding As and HG.

Signes-Pastor et al. (2016a, b) investigated total arsenic and arsenic speciation in rice from the main rice-growing regions of the Iberian Peninsula. The main arsenic species found were inorganic and dimethylarsinic acid. About 26% of commercial rice samples exceeded the permissible As concentration for infant food production as governed by the European Commission.

Medrano et al. (2010) evaluated the association of municipal drinking water arsenic concentrations during the period 1998–2002 with cardiovascular mortality in the population of Spain. Mean municipal drinking water arsenic concentrations ranged from <1 to 118 μg/L. Compared to the overall Spanish population, sex- and age-adjusted mortality rates for cardiovascular (SMR 1.10), coronary (SMR 1.18), and cerebrovascular (SMR 1.04) disease were increased in municipalities with As concentrations in drinking water >10 μg/L.

2.4.3 Portugal

Lousal polymetallic massive sulphide mine is located in the SW Portugal, in the NW region of the Iberian Pyrite Belt, inside of the volcano-sedimentary complex lineament. The Lousal mine is a typical "abandoned mine" with all sorts of problems as a consequence of the cessation of the mining activity and lack of infrastructure maintenance, and heavy metal-enriched tailings have remained at the surface in oxidizing conditions. According to Luís et al. (2011), high concentrations of As occurred within the stream sediments downstream of the tailings sites, up to 817 mg/kg, and acid mine drainage water pH was 1.9–2.9.

In NE Portugal, the Santo António mine is located 3 km northwest of the Penedono village, which was a very important gold and arsenic mining centre in the 1950s. The abandoned waste dumps contain large amounts of arsenic, gold, and silver and medium to great contents of bismuth, copper, lead, manganese, and zinc, while draining waters from tailings have low pH (2.5–4.3), high electrical conductivity, and large concentrations of aluminium, arsenic, sulphate, and other trace elements. According to Madeira et al. (2012), those seepage waters along the dust transported by wind from the tailings, are responsible for the spread of hazardous trace elements, particularly As, to surrounding soils. The mean As concentration in soils in the vicinity of the mine was about 2558 mg/kg. The authors determined the influence of soil amendments (iron oxides, organic matter, and calcium phosphate) on As uptake and translocation by tomato and parsley grown in As-contaminated soil. Results showed that the health risk for adults and adolescents was within acceptable limits with an average weekly intake of 500 g of tomato and 20 g of parsley, although As levels in edible tissues were well above those at uncontaminated control sites.

In a geochemical sampling campaign undertaken by Coelho et al. (2012) in the Panasqueira Mine (Sn-W mineralization) area of central Portugal, an anomalous distribution of several metals and arsenic was identified in various biological samples (blood, urine, hair, and nails from a group of individuals living near the Panasqueira Mine). The results showed elevated levels of As, Cd, Cr, Mn, and Pb as well as elevated potential risk for the health of local residents. Since today the economic exploitation is located in Barroca Grande, Candeias et al. (2014) carried out a study at a S. Francisco de Assis village, located downstream of the Barroca Grande tailings deposit and impoundments, in order to investigate the environmental contamination impact on agricultural and residential soil. Rhizosphere samples, vegetables (*Solanum tuberosum* sava and *Brassica olerácea* L., which constitute an important part of the local human diet), irrigation waters, and road dusts were collected in private residences. The As content in rhizosphere soils exceed 20 times the reference value for agricultural soils (11 mg/kg). The results showed that some edible plants frequently used in the region could be enriched in these metals/metalloids and may represent a serious hazard if consumed. The potatoes tend to have a preferential accumulation in the leaves and roots, while in cabbages most elements had a preferential accumulation in the roots. The inhabitants of S. Francisco de Assis village are probably exposed to some potential health risks through the intake of arsenic, cadmium, and also lead via consuming their vegetables.

The old Mondego Sul uranium mine is located in the western part of the uranium-bearing Beira area, in the Ázere village, near Tábua, Coimbra district. The area belongs to the Central Iberian Zone of the Iberian Massif. The water, stream sediments, and soils in the area were found to be contaminated with U and As (Neiva et al. 2016). Arsenic concentrations were up to 158 µg/L in water, 211 mg/kg in stream sediments, and 223 mg/kg in soils. The authors stated that a restoration of the mining area was necessary to avoid a public hazard.

Marques et al. (2009) investigated the potential of *Rubus ulmifolius*, indigenous to a metal-contaminated site – “Esteiro de Estarreja” – for phytoremediation purposes. The site has a long history of metal contamination. The accumulation of lead, arsenic, and nickel in different sections (roots, stems, and leaves) of the plant was assessed and compared to the metal levels in soil and available fraction. Significant correlations were found among the total levels of Pb and As in soil and levels in plant roots.

Vieira et al. (2011) collected three commonly consumed and commercially valuable fish species (sardine, chub, and horse mackerel) from NE and eastern central Atlantic Ocean in Portuguese waters during 1 year. Maximum mean levels of mercury (0.1715 ± 0.0857 mg/kg, ww) and arsenic (1.139 ± 0.350 mg/kg, ww) were detected in horse mackerel. Based on estimates of noncarcinogenic and carcinogenic health risks, the authors suggested that the analysed species should be consumed moderately due to possible hazard and carcinogenic risks derived from arsenic (in all analysed species) and mercury ingestion (in horse and chub mackerel species).

Marquez-Garcia et al. (2012) analysed the arsenic speciation in soils from São Domingos mine area and found out that arsenate was the major species. The arsenic content and speciation analysis were carried out also in two metal-tolerant species from the area: *Erica andevalensis*, endemic heather from the mining areas of SW Iberian Peninsula, and *Erica australis*, a widely distributed species. The total soil content of As ranged from 194 to 7924 mg/kg, while levels of 1–24.4 mg/k in *E. andevalensis* and 2.7–11.6 mg/kg in *E. australis* were found.

Ereira et al. (2015) sampled sediment, suspended particulate matter (SPM), water and clam *Scrobicularia plana* from a temperate coastal lagoon with anthropogenic impact. Arsenic levels in sediments, SPM, and water presented a spatial concentration gradient. A significant linear regression between arsenic levels in *S. plana* and SPM confirmed that the SPM was the main route for As exposure. Despite the absence of regulatory guidelines, food safety assessment highlighted possible adverse effects of consuming *S. plana* in most contaminated areas.

Signes-Pastor et al. (2016) investigated total arsenic and arsenic speciation in rice over the main rice-growing regions of the Iberian Peninsula. The main arsenic species found were inorganic and dimethylarsinic acid. Samples surveyed were soil, shoots, and field-collected rice grain. Commercial polished rice had the lowest iAs content in Andalusia, Murcia, and Valencia, while Extremadura had the highest As concentrations. About 26% of commercial rice samples exceeded the permissible concentration for infant food production as governed by the European Commission.

2.5 Western Europe

2.5.1 Denmark

Larsen et al. (1998) collected samples of edible mushroom *Laccaria amethystina*, known to accumulate arsenic, from two uncontaminated beech forests, and from the As-contaminated one. Total As values were 23 and 77 μg/g DW in two uncontaminated samples and 1420 μg/g DW in a contaminated sample. The study showed that mushrooms or their associated bacteria, when grown in highly arsenate-contaminated soil (500–800 μg/g), were able to biosynthesize dimethylarsinic acid from arsenic acid in soil. The authors also detected arsenobetaine and trimethylarsine oxide for the first time in *Laccaria amethystina*. Herewith, they recommended not to consume it if collected from contaminated soil due to genotoxic effect of dimethylarsinic acid observed at high doses in animal experiments.

De Gieter et al. (2002) determined levels of As in muscle and liver tissue of 25 sea fish and 4 shellfish species from the North Sea. The highest total As concentrations were found in lemon sole, dogfish, ray, and witch. Their average total As values were above 20 mg/kg WW. The same species as well as the other flatfishes had the highest levels of toxic As (>0.1 mg/kg WW). Toxic fractions (AsTox/AsT%) above 2% were found in sea bass, ling, John Dory, pouting, dab, and brill. It was not observed any preferential concentration in the liver compared to the muscle. The authors emphasized that As content of the North Sea marine food might have reached harmful levels when fish was dried or smoked (0.5 mg/kg WW).

Arsenic occurs naturally in many types of seafood as water- and fat-soluble organoarsenic compounds. According to Taleshi et al. (2010), contrary to water-soluble compounds, the fat-soluble compounds (the so-called arsenolipids) have not been well characterized in the literature. Their paper reported that sashimi-grade tuna fish, with total arsenic content of 5.9 μg/g DW, contained approximately equal quantities of water- and fat-soluble As. The water-soluble As contained predominantly arsenobetaine (>95%) with a trace of dimethylarsinate. The authors isolated and characterized two fat-soluble compounds, which together accounted for about 40% of the lipid-arsenic. This has been the first identification of arsenolipids in commonly consumed seafood.

Epidemiological studies have proved that intake of drinking water with high As levels (>100 μg/L) is associated with risk for cardiovascular diseases. Since studies on lower levels of arsenic have showed inconsistent results, the aim of the study by Monrad et al. (2017) was to investigate the relationship between exposure to low-level As in drinking water and risk of myocardial infarction in Denmark. Arsenic levels in drinking water at baseline addresses ranged from 0.03 to 25.3 µg/L, with the highest concentrations in the Aarhus area. The authors found no overall association between 20-year average concentration of As and risk of myocardial infarction. However, in the Aarhus area, the fourth As quartile (2.21–25.34 µg/L) was associated with an IRR (incidence rate ratios) of 1.48 (95% confidence interval (CI): 1.19–1.83) when compared with the first quartile (0.05–1.83 µg/L). The value of IRR 1.26 (95% CI: 0.89–1.79) was found for ever (versus never) in case of living at an address with 10 µg/L or more arsenic in the drinking water. The authors concluded that their study provides some evidence that even low levels (2–25 µg/L) of As in drinking water might be positively associated with incident of myocardial infarction. However, they found no support for this association at As concentrations below 2 µg/L.

2.5.2 Germany

The dietary intake of arsenic, cadmium, mercury, and lead was studied among young German children with different food consumption behaviour, i.e. own grown foodstuffs vs supermarket products (Wilhelm et al. 2005). The study area comprised an industrialized and a rural area of West Germany. Geometric means of weekly intakes [$\mu g/(kg_{bw} \bullet week)$] were the following: As 1.4, Cd 2.3, Hg 0.16, and Pb 5.3. Geometric mean intake corresponded to the percentage of the provisional tolerable weekly intake (PTWI) as follows: As 9.7%, Cd 32%, Hg 3.3%, and Pb 21%. Arsenic and Hg intake were mainly influenced by fish consumption. The authors pointed out that children from an industrialized area with a substantial food consumption of homegrown vegetables or products from domestic animal products had no increased dietary intake of the analysed elements. Also, health risks due to dietary intakes of As, Cd, Hg, and Pb were low for the children studied.

Floodplain soils of the Elbe river catchment get frequently polluted with metals and arsenic. Among five common floodplain plant species, *Artemisia vulgaris* showed highest concentrations of Cd, Cu, and Hg, As was highest in *Alopecurus pratensis*, while Ni, Pb, and Zn were most elevated in *Phalaris arundinacea* (Overesch et al. 2007). In order to limit harmful transfers into the food chain, the authors suggested that low-lying terraces and flood channels containing highest contamination levels or phytoavailabilities should be excluded from mowing and grazing.

2.5.3 France

Fillol et al. (2010) carried out a cross sectional study to evaluate arsenic exposure of residents living in an area with soil naturally rich in As, through urinary measurements. It was found out that there are significant associations among urinary As concentrations and the consumption of seafood ($p = 0.03$), wine ($p = 0.03$), and beer

($p = 0.001$). The paper showed that the population, living in the study area where soil is naturally enriched in As, is not dramatically overexposed to As. Urinary concentrations of As were close to those of general population and were all far below the regulation values for occupational populations.

The French Nutrition and Health Survey was conducted to determine dietary intakes, nutritional status, physical activity, and levels of various biomarkers for environmental chemicals in the French population (Saoudi et al. 2012). In total, 1500 children and 1515 adults were examined in terms of the sum of inorganic arsenic and its two metabolites, monomethylarsonic acid and dimethylarsinic acid, and for the total arsenic. Arsenic levels observed were similar to or lower than those observed in previous national studies conducted in France and other countries. The main sources of inorganic (toxic) arsenic identified were seafood and wine consumption.

Since seafood, especially fish, is considered as a major dietary source of arsenic, the aim of the study conducted by Sirot et al. (2009) was to assess As intake of frequent French seafood consumers and exposure via biomarkers. The average As dietary exposure was found to be 94.7 ± 67.5 μg/kg bw/week in case of females and 77.3 ± 54.6 μg/kg bw/week in case of males ($p < 0.001$), while the inorganic As dietary exposure was found to be 3.34 ± 2.06 μg/kg bw/week and 3.04 ± 1.86 μg/kg bw/week ($p < 0.05$), respectively. According to the authors, even among high consumers of the seafood, it is not the main source of toxic As.

2.5.4 United Kingdom

Since birds of prey forage over large areas, they might be expected to accumulate contaminants. Erry et al. (1999) tested the hypothesis that As levels in raptors from a region with naturally enhanced As levels were higher than those in birds from an uncontaminated part of Britain. Arsenic residues in kestrels were found to be significantly different between the two groups, but this was not the case for other bird species, and the authors ascribed it to both diet and arsenic metabolism.

Arsenic contamination in Cornwall is widespread due to the historic mining of polymetallic ores and the calcination of ores. In areas such as Camborne/Redruth and Hayle in West Cornwall, arsenic values typically exceed 100 mg/kg in topsoils and occasionally in the subsoil. Camm et al. (2004) investigated the dispersion of As from a calciner stack and from fugitive dusts at the New Mill site (Roseworthy, near Camborne, Cornwall) and found levels of total As above 4000 mg/kg. Agricultural disturbance by ploughing and downslope leaching has probably dispersed the As contamination in the soil. Water samples had As concentrations <50 μg/L, indicating relatively low mobility of soil arsenic. However, the authors emphasized that a large reservoir of arsenic could be remobilized if there was a change in ambient conditions.

Rieuwerts et al. (2006) sampled household dust and garden soil from 20 households in the vicinity of an ex-mining site in SW England and from 9 households in a control village. The results showed clearly elevated As levels, up to 486 μg/g in house dusts and 471 µg/g in garden soils, respectively. Arsenic concentrations in all samples from the mining area exceeded the UK Soil Guideline Value (SGV) of 20 µg/g. No significant correlation was observed between garden soil and house dust As concentrations. The most important outcome of the study is the fact that it supports the concerns expressed by previous authors about the significant As contamination in SW England and the potential implications for human health.

2.5.5 Ireland

The study by McGrory et al. (2016) amalgamates readily available national and subnational scale datasets on groundwater As in the Republic of Ireland. Several arsenic databases were integrated and the data modeled using statistical methods appropriate for non-detect data. In addition, geostatistical methods were used to assess principal risk components of elevated arsenic related to lithology, aquifer type, and groundwater vulnerability. For the majority of sampled locations in the study, the As levels in groundwater is below both the Irish GTV value of 7.5 µg/L and the WHO and USEPA value of 10 µg/L. Only a small number of locations exhibit elevated As concentrations due to Silurian and Ordovician metasedimentary formations. The study provides the preliminary steps towards the creation of a national database on Irish groundwater As levels. According to the authors, the presence of regional hotspots of contamination warrant further detailed investigations.

3 Australia

Arsenic is commonly found throughout Australia, and localized As contamination problems have been reported in many of the states and territories. The mining industry has contributed widely to the incidences of arsenic contamination throughout this continent. Not only has the mining of mainly gold deposits enhanced the release of As into the environment but the mining processes associated with gold extraction also left a lasting legacy of an As-contaminated environment.

Up to date, residual arsenic contamination from historical gold mining activity persists in the goldfields region of Victoria where elevated arsenic concentrations have been observed in mine waste and some residential soils, surface, and groundwaters. In 1991, Department of Manufacturing and Industry Development (1991) of Australia reported extremely variable As concentrations in surface and groundwater in many areas of rural Victoria, ranging from <0.001 to 2.83 mg/L. In comparison, surface water from the greater Melbourne region contained As concentrations between <1 and 52 µg/L (DMID 1991). Hinwood et al. (1999) reported a similar range of As concentrations, from 1 to 5000 µg/L, in surface and groundwater of rural Victoria.

However, most Victorian households currently depend on a reticulated water supply, whereas self-extraction from rainwater tanks or from surface or groundwater contributes less than 4%. Study of Hinwood et al. (2003) also reported high variability of arsenic concentrations in drinking water and soil in this region, ranging from below the detection limits to 73 μg/L and from 1.7 to 9900 mg/kg, respectively. Past consumption of contaminated water was possibly even greater, but their study revealed that residents living on the soil of high arsenic content (up to 9900 mg/kg) consume water with arsenic concentrations ranging from below the detection limit to 1.3 μg/L. Contrary, participants consuming drinking water with the highest concentration of arsenic were those living in the area where arsenic concentrations were lowest in the soil.

The small-area ecological study conducted by Pearce et al. (2012) on a population of residents in 61 contiguous statistical local areas in Victoria identified a small but significantly increased risk of all cancers combined, prostate and breast cancers, melanoma, and chronic myeloid leukaemia. Their study, however, revealed that arsenic-contaminated drinking water is unlikely to be the major contributor to arsenic exposures for studied population.

Although mining activities have contributed to the contamination of soil and water primarily in the Western Australia and Victoria, the same can be found in other parts of the Australian territory as well. For instance, in the upper part of the Macleay River catchment, northern New South Wales (NSW), historic mine waste disposal practices of the Hillgrove mineral field, a major producer of gold (Au) and antimony (Sb), have resulted in an As- and Sb-contaminated sediment dispersion plume. This contamination extends few hundred kilometres eastwards to the coastal floodplain at Kempsey, where population density is higher and land use more intense. Due to the production and subsequent redistribution of sediment from the upper catchment, the surface soils of the Macleay River floodplain are enriched with both arsenic (As) and antimony (Sb) (up to 40 mg/kg of both metalloids).

Other mining processes associated with the recovery of rare metals such as titanium also result in the arsenic contamination of groundwater systems. Binning et al. (2001) reported that mining of titanium from sand beds near Newcastle, New South Wales, led to oxidation of arsenopyrite material in the sand beds which subsequently led to an increase in As concentrations up to 30 mg/L in the unconfined aquifer.

Other anthropogenic activities such as agriculture, forestry, and industry have also contaminated soil and water at a localized scale in Australia. Namely, arsenic has been widely used in the agricultural industry, as both a pesticide and herbicide. Industrial activities, such as timber treatment plants and ammonia production processes, also led to occurrences of elevated As levels in surface and groundwater. Throssell and Blessing (2001) reported As concentrations in the range from 0.005 to 220 mg/L in groundwater accidentally contaminated by As solution used in the production of ammonia at Kwinana, Western Australia.

The geological setting combined with an increase in population density and consequently increased water requirements is becoming one of the main reasons for occurrences of elevated arsenic concentrations in groundwater in Australia. Smith and Jankowski (2001) identified elevated concentrations of As (up to 300 μg/L) in groundwater used as a source of drinking water in the small coastal community of Stuarts Point, New South Wales. According to these authors, the source of As contamination of the groundwater from Stuarts Point was due to a number of processes controlling the release of arsenic into the groundwater environment, with the desorption of As from Al hydroxides and As-enriched Fe oxyhydroxides and the oxidation of arsenical pyrites as the dominant one. Similarly, McLean and Jankowski (2001) attributed elevated As concentrations (up to 70 μg/L) in groundwater collected from the lower Namoi River catchment, New South Wales, to natural mobilization of As from within the groundwater environment.

The combination of increasing population density in urban areas and low rainfall increased groundwater abstraction and contributed to the emergence of As enrichment within the Gwelup groundwater management area, several kilometres north of the Perth city centre. Namely, a prolonged drought in Perth is causing the water table to fall in some parts of the metropolitan area to the point where substantial changes in the chemistry of groundwater are taking place, including acidification and the release of As and metals from aquifer sediments. According to study of Appelyard et al. in 2006, groundwater in the Gwelup groundwater management area in Perth has been enriched in As (up to 7000 μg/L) due to the exposure of pyritic sediments caused by reduced rainfall, increased groundwater abstraction for irrigation and water supply, and prolonged dewatering carried out during urban construction activities. Arsenic was initially released into groundwater through the oxidation of arsenian pyrite when sulfidic peaty sediments were dewatered and excavated for the construction of housing estates. These activities caused the pH of shallow groundwater to decline from 6–7 to as low as 2.5 and produced arsenic concentrations of up to 22,000 μg/L in groundwater. Although the pH of groundwater was subsequently moderated (up to 5.5), arsenic concentrations remain high in many areas (up to 800 μg/L), posing an ongoing health risk to residents who use domestic bores for garden watering and other household uses.

According to Appleyard et al. (2006), decline in the water table due to prolonged period of low rainfall, and the disturbance of sulfidic peat soils by dewatering and excavation in the Perth suburb of Stirling, led to widespread acidification of groundwater at the water table in the residential area and contamination of groundwater by arsenic and other metals. The dewatering and peat excavation caused pH values to drop as low as 1.9, resulting in a high concentration of arsenic (up to 7 mg/L), aluminium (up to 290 mg/kg), and iron (up to 1300 mg/kg) in the groundwater. Although acidic water extends 5–10 m below the water table and the deeper groundwater remains unaffected by contamination, the fact that groundwater forms 70% of Perth's total water usage and sulphide-rich peat soils are common in the region brings many people at risk of consuming water contaminated with arsenic.

Historically, much of this arsenic contamination has been isolated due to the small population base of Australia, but the current changing demographics of Australia and an increased concern about the environment put forefront As contamination as an important public issue in this country. Recently, a lot of attention was given to elevated levels of arsenic in food, primarily in rice and rice-related products.

Williams et al. (2006) reported total As (tAs) concentrations in Australian rice in the range from 20 to 40 μg/kg, with a mean of 30 μg/kg ($n = 5$), whereas Juhasz et al. (2006) reported 189 ± 18 μg/kg of As in Australian long-grain white rice. Rahman et al. (2014) studied total and speciated As in Australian-grown and imported rice on sale in Australia. According to their study, the mean and range of tAs concentrations in Australian-grown rice were 270 μg/kg and 188–438 μg/kg, respectively. The highest tAs was found in organic brown rice (438 ± 23 μg/kg) followed by medium-grain brown rice (287 ± 03 μg/kg) and organic white rice (283 ± 18 μg/kg). It is notable that tAs concentration in the organic brown rice exceeds the FAO/WHO (1985) recommended maximum permissible limit of 300 μg/kg. In general, total As concentrations in Australian-grown rice were higher than in imported rice on sale in Australia, with the exception of Italian Arborio rice. While Asian rice contained mainly inorganic As (iAs; 86–99%), 18–26% of the tAs in Australian-grown rice was dimethylarsinic acid (DMA). Among the Australian-grown rice varieties, the highest concentration of organic arsenic, in the form of dimethylarsinic acid (DMA), was found in organic brown rice (115 ± 02 μg/kg), which was ~26% of the tAs. Since inorganic arsenic presents a significant fraction (about 63%) of tAs in this rice type, Australian organic brown rice may be a matter of human health concern, especially for infants who eat cereals and formulas manufactured from organic brown rice and rice bran and for adults who regularly eat organic brown rice.

Islam et al. (2017) investigated total and inorganic arsenic (As) content in rice and rice-based diets obtained from supermarkets in Adelaide, South Australia, in 2015. Results of their research show that of the 59 rice-based products, 31 (53%) had As higher than the maximum level of 100 μg/kg recommended by European Union (EU) for young children and 13 (22%) samples had As higher than maximum level of 200 μg/kg recommended for adults (EFSA 2014). Arsenic content varied in order rice crackers > baby rice > rice cakes > puffed rice > other rice-based snacks > ready-to-eat rice. Of the six studied categories of rice-based products, except ready-to-eat rice, all others exceeded the value recommended by EU for young children. Even manufacturers recommended servings deliver significant amounts (0.56–6.87 μg) of inorganic As.

4 New Zealand

Similarly to Australia, arsenic presents one of the major national groundwater quality issues in New Zealand. While nitrate contamination is a major issue in shallow groundwater due to numerous anthropogenic sources, naturally elevated concentrations of arsenic, iron, and manganese are found in deeper groundwater. In Canterbury, on the other hand, high arsenic, iron, and manganese concentrations are often encountered in areas of reduced groundwater. In this region, the elevated As, Fe, and Mn levels in groundwater originate from minerals in the aquifer sediments, which are mostly derived from the weathering of greywacke rock. Furthermore, weathering of arsenic-bearing source rocks in the mountains leads to accumulation of arsenic in fine-grained estuarine deposits near the coast. Reduced groundwater conditions make the arsenic more mobile and limit arsenic being adsorbed by iron oxides, which are also soluble in such conditions. Occasionally, high arsenic may originate from old sheep dips or wood treatment plants. Consequently, at a number of sites, mostly in the Woodend and Sefton areas, groundwater exceeds the Provisional Maximum Acceptable Value (PMAV) for arsenic (10 μg/L).

In a review of contaminants in New Zealand drinking water, Davies et al. (2001) identified arsenic concentrations exceeding half-PMAV (5 μg/L) in 70 distribution zones serving a population of approximately 285,000 and those exceeding the PMAV (10 μg/L) in 28 distribution zones serving a population of approximately 21,000. According to these authors, higher-than-average arsenic occurs in greywackes (old ocean sediments), schists (the same sediments transformed with heat and pressure and chemically similar to greywackes), Tertiary volcanics, and some coals and peats. Geochemical interactions between the water and mentioned geological units, i.e. mineral dissolution and weathering, sorption/desorption processes, and leaching, cause an increase in arsenic concentrations in associated groundwater. Rates of weathering, dissolution, and leaching of various major and trace elements are further increased by geothermal heating in this region.

In most of the previous work on arsenic in the Waikato region's water supplies, the focus was on arsenic in surface waters and, in particular, the Waikato River. The Waikato River currently receives a significant load of geothermal arsenic and its concentrations equal twice the drinking water standard before treatment. According to the Environment Waikato Technical Report (2006), arsenic concentrations in the Waikato River decreased over the years due to reducing inputs from the Wairakei Geothermal Power Station. Removal of arsenic during water treatment at Hamilton has also improved, with the efficiency of 90% of arsenic removal attained in 2002, resulting in the average arsenic concentration of Hamilton drinking water in 2002 of about 2.3 μg/L.

5 Africa

High concentrations of arsenic have been recorded in both surface and groundwater in different parts of Africa, including Botswana, Burkina Faso, Ethiopia, Ghana, Morocco, Nigeria, South Africa, Tanzania, Togo, and Zimbabwe. Although the elevated level of arsenic in groundwater has been documented only sporadically across the continent, it represents a serious health threat in many African countries that are already facing multiple challenges related to water quality and shortages.

Ahoulé et al. (2015) summarized findings regarding arsenic distribution and sources in African waters and probable extent of As contamination in Africa. The values quoted in their work show a very large range of values, from 0.02 to 1760 μg/L for groundwater and up to 10,000 μg/L for surface water. According to the African literature, high level of arsenic in surface water is generally related to mining operations, agricultural drains, local sediments, disposal, and incineration of municipal and industrial wastes, whereby mining activities remain the main source of surface water pollution. As for groundwater, arsenic occurrences are generally related to local geology, mineralization, geothermal waters, etc. Since African people rely mainly on local groundwater sources for their water needs, either deep boreholes or shallow wells, the use of such water over a long period of time can lead to fatal consequences.

Botswana is presently one of only a few well-studied cases of geogenic arsenic contamination on the African continent. Huntsman-Mapila et al. (2006) reported elevated levels of arsenic (up to 116 µg/L) in deep groundwater (>70 m) southeast of the Okavango Delta, near the town of Maun, while later study of Huntsman-Mapila et al. in 2011 recorded even higher arsenic concentrations in shallow groundwater underlying the Camp Island (up to 3.2 mg/L). Mladenov et al. (2013) also determined elevated levels of arsenic (up to 180 µg/L) in the groundwater of the Okavango Delta. According to these authors, high arsenic zones in this large arid zone wetland are probably controlled by evapotranspiration and throughflow conditions of the aquifers. High evapotranspiration rates possibly concentrate As and other solutes, while alkaline pH leads to desorption of arsenic or dissolution of arsenic sulphides and formation of thioarsenic complexes which keep arsenic in solution.

In Central Burkina Faso, where surface water availability is limited, groundwater is the main source of water supply for the local communities. In contrast to groundwater availability, groundwater quality has received little attention in Burkina Faso, and the available data indicate that groundwater resources are frequently exposed to various sources of contamination. Few studies, including the one from Smedley et al. in 2007, have reported the occurrence of high arsenic concentrations (>10 µg/L) in groundwater around gold mineralized zones in the north-central region of Burkina Faso. In places, Smedley et al. (2007) found extremely high arsenic concentrations in the groundwater in Burkina Faso (up to 1630 µg/L), while Kusimi and Kusimi (2012) reported a similar range of concentrations (up to 1760 µg/L) in groundwater in Ghana. In both countries, occurrences of high arsenic levels in groundwater were attributed to the underlying geology. According to Bretzler et al. (2017), 14.6% of Burkina Faso rural population uses drinking water from boreholes containing arsenic concentrations higher than the maximum permissible levels (>10 µg/L). The arsenic in this region derives from zones of gold mineralization in Birimian (Lower Proterozoic) volcano-sedimentary rocks and altered sulphide minerals (pyrite, chalcopyrite, arsenopyrite).

Nonetheless, mining activities in Ghana seem to affect more surface waters than the groundwater. Namely, study of Asante et al. (2007) conducted in Tarkwa, capital of Tarkwa-Nsuaem Municipal district (South Ghana) and the centre of gold mining, revealed much higher arsenic concentrations in surface waters (0.5–73 µg/L) compared to the groundwater (<0.1–4 µg/L). A similar observation was reported by Smedley (1996) for Obuasi, another gold mining area in Ghana, where As concentrations ranged from <2 to 175 µg/L and from <2 to 64 µg/L in surface waters and groundwater, respectively. Serfor-Armah et al. (2006) measured even higher arsenic concentrations in the surface waters in Prestea in Ghana, ranging from 150 to 8250 µg/L, whose origin was also linked to mining activities in the area.

Hadzi et al. (2018) assessed the contamination and health risk of heavy metal(loid)s, including arsenic, in selected waterbodies around gold mining areas in Ghana. They found maximum As concentrations (0.23 ± 0.03 mg/L) during a dry season. Compared to the pristine areas encompassed by their study, arsenic concentrations in rivers of the mining areas were two to three orders of magnitude higher. Although there could be other sources accounting for heavy metal(loid) presence in these rivers, anthropogenic activities, primarily mining, are suspected to be the major contributor.

Despite the apparent differences in the concentration of arsenic between surface and groundwater in Ghana, the same high concentrations of arsenic in urine samples among residents of both the mining town of Tarkwa and the town of Accra (devoid of any mining activities) have been reported, suggesting that arsenic contamination has another source, probably food. However, drinking water is still considered a significant pathway of human exposure to arsenic.

In Ethiopia, in the East African Rift Valley, Reimann et al. (2003) found arsenic in the concentration higher than 10 μg/L in 9 out of 138 wells (i.e. 6.5%). For comparison, in Botswana, 6 out of the 20 wells used for the analysis of arsenic showed concentration exceeding the WHO recommended limit (10 μg/L). Studies of Rango et al. (2013) reported high arsenic concentrations in groundwater of the main Ethiopian Rift aquifers, up to 278 μg/L, while surface water of the region exhibited even higher arsenic concentrations (up to 566 μg/L). While elevated levels of arsenic in surface water in Ghana are generally attributed to mining activities, local sediments are considered the main source of arsenic in water in the Rift Valley, Ethiopia.

In Southern Nigeria, study of Asubiojo et al. (1997) reported arsenic levels in the groundwater ranging from 0.4 to 6.88 μg/L, whereas sporadic occurrences of elevated As concentrations (up to 28 μg/L) were reported sporadically near mechanic and panel beaters workshops.

Rezaie-Boroon et al. (2011) have also highlighted high concentrations (6460 μg/L) of arsenic in the surface water in the vicinity of Lomé, Togo, and other big cities. High concentration of arsenic in surface waters near abovementioned cities was attributed to the impact of the effluents from the industrial activity as well as hazardous waste dumping.

Nonetheless, borehole water is a primary source of drinking water supply in many rural parts of Africa, including South Africa. In the Limpopo Province, groundwater accounts for about two-thirds of the total water supply. The elevated arsenic concentrations have been primarily related to bedrock geology and Au mineralization. In places, where As concentrations reach 1000 μg/L in groundwater, cases of severe arsenic poisoning have been reported. Another source of arsenic to groundwater and soil in South Africa is the use of chromated copper arsenate in the preservation of timber. Although arsenic protects the timber from wood-destroying insects, improper disposal of waste from timber plants contaminates the soil and water systems around the plants. Kootbodien et al. (2012) detected high levels of arsenic in school vegetable garden soils as well as in vegetables grown in these gardens in Johannesburg area of South Africa, whereas George and Gqaza (2015) reported elevated arsenic levels (1.5–1.9 mg/kg) in leafy vegetables collected from home gardens in the Eastern Cape Province of South Africa.

6 Russian Federation

About 70% of the population of the Russian Federation receives drinking water from surface water sources, 40% of which do not comply with hygienic standards, of sanitary or aesthetic nature. Permafrost which occupies about 65% of Russian territory (including the whole Arctic and the bulk of Siberia and the Far East) is the main cause of the rare use of groundwater sources in the northern regions of Russia.

In general, the quality of drinking water in Russia was reported to be low due to poor aquifer protection from surface contamination, lack of sanitary protection, and delayed repair, cleaning, and disinfecting of wells and interception ditches. The lack of municipal financing led to serious deterioration of water distribution and sewerage networks, as well as numerous accidents on those networks that led to secondary pollution of drinking water; and instead of systematic preventive maintenance and repair of water supply facilities and networks, problems are usually addressed after the accidents. According to the study of Dudarev et al. (2013), about 28% of the Russian population currently consumes highly mineralized drinking water (1.610 g/L), and about 50 million people in the country (one-third of the population) consume drinking water with enhanced iron content.

The literature on arsenic occurrences in the ground- and drinking water in the territory of Russian Federation is scarce and related to sporadic occurrences of elevated levels of As, associated with both the geogenic and anthropogenic sources. The arsenic limit in drinking water and reservoirs for domestic use recommended by Russian Ministry of Health is 10 μg/L, same as the limit recommended by the World Health Organization.

Among some of the main causes of high arsenic in groundwater in Russia, arsenic-containing ore tailings represent a powerful source of technogenic arsenic. Yurkevich et al. (2012) reported arsenic concentrations substantially greater than the maximum permissible water-use standards (10 μg/L) in drainage waters from tailings at the Belovo (Kemerovo region, Russia) and Karabash (Chelyabinsk region, Russia) ore processing facilities. These drainages enter the Bachat and Sak-Elga Rivers elevating levels of dissolved arsenic up to 21 μg/L and 12 μg/L, respectively, i.e. above values considered safe for use. Further studies identified zones of geochemical anomalies near the Belovo Zn-processing and the Karabash mineral-processing plants where the concentrations of As, Fe, Cu, Zn, Cd, and Pb were two to three orders of magnitude higher compared to the drinking water standards (for rivers) and background levels (for snow).

Chelyabinsk and its region are nowadays one of the most polluted subjects of the Russian Federation, whereas Karabash is considered as one of the most polluted cities in the world. The high rate of pollution in this city occurs due to the presence of the copper smelter, contributing also to an increased As emissions into the environment. As previously mentioned, toxic elements migrate out of numerous tailing dumps with drainage streams that discharge into the nearest rivers and also seep into groundwater. Concentrations of dissolved metals, As and Sb, in drainage waters and influenced rivers often exceed drinking water standards and background levels by one to four orders of magnitude.

In turn, as confirmed by studies of Gilmundinov et al. (2014), people living in Chelyabinsk area were characterized by different health problems like impaired nervous and haematopoietic systems functioning in children and a high number of pregnancy complications, premature births, and neonatal morbidity. Study of Skalny et al. (2016) has shown that whole blood As level in children living in Karabash significantly exceeds that in two other cities of Chelyabinsk region, Varna and Tomino by 69% and 57%, respectively. In particular, hair As in children living in As-polluted region was nearly 34-fold higher as compared to the respective values from Varna and Tomino.

High levels of arsenic in groundwater associated with mine tailings were also registered by Bortnikova et al. (2018) in the vicinity of the Komsomolsk and Berikul gold deposit, located in the Kemerovo region. While most of the element (Fe, Pb, Cd, Cu, Zn, Se, As, Sb, etc.) concentrations in the Komsomolsk tailings pond (pH ~8) do not exceed the maximum permissible concentrations (MPC), the latter does not apply to arsenic and antimony, whose concentrations exceed the MPC by factors of 21 and 170 on average, respectively. The element concentrations in the Berikul ponds are even higher than those at the Komsomolsk tailings pond. The ponds that formed on the Berikul mine tailings have acidic (pH ~2) and extra-mineralized solutions (up to 100 g/L) with extremely high As concentrations for any type of water worldwide influenced by the mining industry, from 10^4 to 10^7 μg/L. Surface drainage from the tailings enters the Voskresenka River and the concentrations of As (~8 μg/L) in the river water approach the MPCs established by the Russian Ministry of Health (10 μg/L). According to Bortnikova et al. (2018), the uncontrolled leakage of acidic and highly mineralized solutions through a natural geological fault into groundwater horizons causes groundwater contamination. The latter was confirmed by increased arsenic concentrations in the water from a well located near the fault (up to 89 μg/L).

For the European part of Russia, on the other hand, there are no reports of groundwater contamination with arsenic. Salminen et al. (2004) observed elevated concentrations of arsenic in surface waters of the Barents region. However, the risk of geogenic groundwater contamination to human health in the Barents region is probably very small since people do not depend on groundwater as a source of drinking water.

Similarly to the Barents region, the geological setting is responsible for elevated levels of arsenic in groundwater in Dagestan. Namely, regional pollution of fresh groundwater with arsenic was attributed by Kurbanova et al. (2013) to assemblages of arsenic minerals in some sandy-clayey beds in contact with productive horizons. Intensive exploitation of artesian waters and related activation of the groundwater movement have led to an increase in heat and mass exchange between fluids and mineral particles of rocks, which also facilitated the increase in arsenic concentration in associate groundwater. In places, arsenic in the groundwater exceeds 40 times the WHO limit. Moreover, unlike the Barents region, 65% of the need for public water supply in Dagestan is provided by artesian groundwater.

Elevated levels of arsenic are also found in river waters nearby geothermal fields. One such occurrence was described by Ilgen et al. (2011) for Falshivaia River, situated within the Mutnovsky geothermal region on the Kamchatka peninsula. This high-temperature field is used for electricity production, whereby the spent fluids, containing elevated concentrations of arsenic (up to 9.3 mg/L), are discharged into near-surface environments. Fortunately, the extent of elevated arsenic concentrations in surface water is limited by adsorption to the bottom sediment and dilution, and geothermal waste fluids released in the river create only a localized area of arsenic contamination. However, one of the wells was the subject of hydrothermal explosions in 2003–2004 and, as reported by Melnikov (2004), has been freely discharging hot fluid (that enters the Falshivaia River) since then.

According to Zakharova et al. (2002), fertilizer industry plants, and particularly the resulting solid waste storage areas, represent one of the main potential sources of environmental contamination in Russia with regard to arsenic. Built in the 1950–1960s, without necessary technical measures for prevention of environmental pollution, solid waste from fertilizer plants is neither processed nor treated in Russia but dumped for storage. Consequently, soils in areas near fertilizer industry plants often contain elevated levels of arsenic, which is unfortunately reflected further in elevated arsenic levels in cultures grown on these soils, as well as in the groundwater of that area. Total arsenic content in soils around the Voskresensky phosphorus fertilizer plant in the Moscow Region ranged from 0.51 to 1.65 mg/kg, while plants grown on these soils (fruits, vegetables, grains) contained from 0.41 to 1.32 mg/kg of total arsenic. The health risk assessment study conducted by Zakharova et al. (2002) revealed that the arsenic exposure pathways through “ingestion of meat” and “inhalation (indoor/outdoor)” pose a negligible health risk, while arsenic exposure pathways through “ingestion of agricultural products”, “groundwater uptake”, and a minor extent “dermal contact” and “direct soil ingestion” appeared significant.

7 Central, Western, and Far East Asia

The water system of the Aral Sea Drainage Basin covers the main part of Central Asia. Spreading of persistent pollutants in this spatially extensive water system was investigated by Törnqvist et al. (2011). Overexploitation of water caused a significant lowering of the river discharge into the Aral Sea, making groundwater flows increasingly important for the overall water budget. Agricultural and industrial sources distributed throughout the water system area are considered as the main sources of pollutants, including arsenic, producing a cumulative health hazard in the downstream surface waters. The high spatial variability of concentrations of As in the river water is considered to result from its local presence in the top soil of the agricultural fields.

7.1 Iran

The arsenic-related groundwater problems in Iran are mostly from naturally occurring sources. Barati et al. (2010) found that in drinking water sources of 21 out of the 530 studied villages from cities Qorveh and Bijar, the level of As (and Cd and Se) exceeded the WHO or the National Standard limits. The concentration of arsenic in the drinking water ranged from 42 to 1500 μg/L, and signs of chronic arsenical poisoning were found in 180 of 587 investigated participants from the affected areas (Mee's line, keratosis, and pigment disorders). A strong linear relationship between arsenic exposure and occurrence of multi-chronic arsenical poisoning was established.

Keshavarzi et al. (2011) investigated the elevated As content in the groundwater of the Kurdistan and West Azerbaijan provinces in the western Iran, the main sources of drinking water. The travertine springs, used for bathing, were also studied as a possible source of arsenic in groundwater. The total arsenic in travertine springs ranged from 212 to 987 μg/L and in the groundwater from 0.4 to 689 μg/L. Arsenite was the dominant arsenic species in the travertine spring (68.2–98.9%). The geochemical investigation confirmed that the travertine springs, belonging to a hydrothermal system related to volcanic setting of the investigated area, was the main source controlling arsenic concentration in the groundwater. Further investigation focused on the occurrence and distribution of As in other environmental samples from this area. Specifically, in addition to drinking water, the agricultural soil, the alfalfa hay used for sheep and blood and wool samples from sheep were investigated by Keshavarzi et al. (2015). Sampling was performed in the Ebrahim-abad and Babanazar villages in the Kurdistan province (western Iran). Increased levels of As were found in all investigated samples. The total As concentrations ranged from 119 to 310 μg/L in the drinking water, 46.7–819 mg/kg in soil, 1.90–6.90 mg/kg in vegetation, 1.56–10.8 mg/kg in the sheep's wool, and 86.3–656 μg/L in the blood samples. The most probable source of As in the sheep samples were the drinking water and plants used for their feeding. The signs of long-term exposure to inorganic As, liver damage and anaemia, were observed in sheep. The obtained results raise concern about possible human exposure to increased arsenic levels through the food chain.

The Sahand region reservoir in the north-western Iran (East Azerbaijan province) is an important reservoir of drinking water for the region, as well as water used for industrial and agricultural purposes. Mosaferi et al. (2017) found elevated concentrations of arsenic (up to 1440 μg/L and a mean concentration 172 μg/L) in the water basin and dam. The contamination is especially significant in the Almalu River and inside the Sahand reservoir. Regional geological formations and volcanic activities are considered as the main sources of the natural genesis of arsenic in the study area.

The canned tuna fish in Iran as a possible source of human exposure to arsenic was studied by Rahmani et al. (2018). A review and a meta-analysis of metal concentrations were performed and carcinogenic risks were evaluated. While concentrations of As were below the recommended limits, according to the Incremental

Lifetime Cancer Risk (ILCR) for As (3.21E-5 in adults and 4.18E-5 in children), it was concluded that adults and children, consuming canned tuna fish in Iran, have a carcinogenic risk due to As.

As a main pistachio producing country, water and five commercial pistachio cultivars from four geographical regions of Iran were analysed with respect to arsenic and other heavy metals by Taghizadeh et al. (2017). The infield metal content in soil showed good correlation with that of pistachio. The highest level of arsenic was determined in the Kaleghoochi cultivar (mean concentration 1.963 ± 0.005 mg/kg) and Sarakhs region. However, the pistachio samples were found to be safe for consumption.

Additional information on arsenic concentration in various food products was provided by Hashemi et al. (2017) in a review of 40 studies investigating level of heavy metals contamination in food in Iran. Kelishadi et al. (2018) conducted a randomized 8-week clinical trial with the aim to analyse human milk with respect to several metals, including arsenic, and investigated influence of the jujube fruit consumption in reducing the concentration of the investigated metals. The study included 40 postpartum mothers in Isfahan, which is the second largest and polluted city in Iran. The mean (standard deviation) concentrations of arsenic were 1.23 (0.63) µg/L, while its concentration declined in both groups, as for other metals. The consumption of jujube fruit exhibited no significant change on the arsenic level.

7.2 Kuwait

Seafood is an important part of diet in the Arabian Gulf countries. Husain et al. (2017) studied the speciation of As in fish, shrimp, and crab in 578 samples of commonly consumed seafood, of 15 different species, from the main fish markets in Kuwait. The mean daily intake of inorganic As through fish consumption was 0.058 µg/kg/day, and the 95th percentile was 0.15 µg/kg/day. While the mean intake level did not exceed the incremental lifetime cancer risk (ILCR) at 1 × 10–4, the 95th percentile of inorganic As intake showed an ILCR of 2.7 × 10–4. A higher mean intake of inorganic As was estimated for the Kuwaiti children (aged 6–12 years), 0.10 µg/kg/day with an ILCR of 1 × 10–4. The hamour fish (*Epinephelus coioides*) was found to be the main source of inorganic As intake. Arsenobetaine was the dominant As species in the tissues of all seafood samples.

7.3 Oman

The concentrations of arsenic in the marine biota (fish and various bivalves) and coastal sediment from the Gulf and Gulf of Oman (Bahrain, Oman, Qatar, and the United Arab Emirates (UAE)) were investigated by deMora et al. (2004). In the investigated sediment samples concentrations of As were low (0.7–9.6 µg/g).

However, high concentrations of As were measured in clams and pen shells, 156 μg/g and 153 μg/g, respectively. The As concentrations in the muscle of the orange spotted grouper *(Epinephelus coioides*, hamour) and the spangled emperor (*Lethrinus nebulosus*, sheiry) varied between 0.83–14.4 and 2.5–10 μg/g, respectively.

7.4 Arabian Gulf

Kosanovic et al. (2007) analysed life essential and toxic elements, including As, in liver and muscle samples of red spot emperor (*Lethrinus lentjan*) from three different locations of the Arabian Gulf Coast. Despite the determined increased content of metals in highly industrial areas, the values did not exceed permitted levels, and fish was considered safe for human consumption.

Levels of total arsenic and arsenic species in marine biota (clams, pearl oyster, cuttlefish, shrimp, and seven commercially important finfish species) in the western Arabian Gulf were analysed by Krishnakumar et al. (2016). The total As concentrations in bivalves ranged from 16 to 118 mg/kg, dry weight, but the inorganic As contributed on average less than 0.8% of the total As, while arsenobetaine formed around 58%. In the remaining seafood (cuttlefish, shrimp, and finfish), the total As concentrations ranged from 11 to 134 mg/kg, dry weight, and the inorganic As and arsenobetaine contributed on average 0.03% and 81% of the total As, respectively. No significant relationship was found between tissue concentrations of the total and the inorganic As in the investigated samples.

7.5 South Korea

The Gubong mine, once among the largest Au-Ag mines in Korea, was the source of arsenic contamination for the surrounding area. Woo and Choi (2001) analysed surface water from the nearby Guryong-chun stream, groundwater from domestic wells tapped into the floodplain deposits along the stream and seepage from the mining wastes. In said waters, the levels of As, Cd, and Mn exceeded the drinking water guidelines of WHO, and the highest concentration of As, 0.167 mg/L, was determined in the floodplain area. The source of arsenic in the water was arsenopyrite, and its concentration was controlled through adsorption-desorption processes with iron oxyhydroxides and solubility of carbonate minerals and is strongly pH dependant. The arsenic level in surface water reflected its concentration in the stream sediments.

Hong et al. (2018) studied the speciation of arsenic in water, suspended particles, zooplankton, sediments, and sediment porewater from the freshwater and saltwater region of the Youngsan River Estuary, in South Korea. The freshwater samples showed significantly lower values of arsenic (mean concentration 1.5 μg/L) compared to the saltwater (mean concentration 5.2 μg/L), but the contrary was found for suspended particles. The major form in water and particle samples was As(V). The results suggested that direct consumption through the food web plays a considerable role in the bioaccumulation of As in the zooplankton, with As(V) and As(III) as the dominant forms. However, the levels of As in the Youngsan River Estuary did not exceed the tolerable levels of ecotoxicological risk.

Hong et al. (2014) investigated water, sediment, and biota in the Pohang City area significantly influenced by the surrounding industry. Arsenic was found in all investigated aquatic organisms, with largest mean concentration determined in crab, than bivalves, shrimps, and gastropods, while fishes exhibited the lowest concentrations. It was found that the concentrations of As in the investigated biota, especially in fishes, and filter-feeders, were dependent on its concentration in the water. The bioaccumulation factors suggested, however, that As does not biomagnify in either freshwater or marine food webs. Arsenobetaine was the dominant form in fishes, bivalves, crabs, and shrimps and As(III) in the freshwater snails.

Jung et al. (2018) investigated arsenic contents in different types of rice commonly consumed in Korea (white, brown, black, and waxy rice) and microwavable ready-to-eat rice products. Mean content of inorganic As in the ready-to-eat rice products was 59 μg/kg (dry weight basis), with the range of 20–131 μg/kg. While in the investigated ready-to-eat rice products inorganic As was below the legal maximum level (200 μg/kg) for adults, 17% of products was not appropriate for the infant and young children foods (the inorganic As maximum level 100 μg/kg set by European Union). In the white (polished) rice, the determined mean value of inorganic As was 65 μg/kg, with the range 33–120 μg/kg, and among 51 different samples, two contained inorganic As over 100 μg/kg. The mean inorganic As content in the brown rice was 109 μg/kg, with the range 67–156 μg/kg, and 70% of samples were found not suitable for production of foods for infants and young children. The non-polished brown rice contained significantly higher content of inorganic As than the polished rice. The mean inorganic As content in the unpolished black rice was 91 μg/kg, with the range of 22–148 μg/kg, and 5 out of 14 tested black rice samples had inorganic As level over 100 μg/kg. Brown and black rice showed significantly higher quantity of inorganic As compared to the polished non-waxy white rice and polished waxy rice. The mean inorganic As content in the polished waxy rice was 66 μg/kg, with the range 46–85 μg/kg. None of the waxy rice samples contained over 100 μg/kg of inorganic As. On the other hand, brown and black rice contained significantly higher content of inorganic As than white rice and waxy rice.

7.6 Japan

Sources of arsenic in rivers of Japan are mostly natural, resulting from geothermal activities and dissolution from soil and sediment. Miyashita et al. (2009) investigated levels of arsenic in water and biological samples of the Hayakawa River (Kanagawa, Japan), where it is delivered from hot springs at Mt. Hakone. Arsenate prevailed in the hot spring water, river water, and the water bug from the river, while oxo-arsenosugar-glycerol and/or oxo-arsenosugar-phosphate were determined in the green macroalgae, crustaceans, and several fish samples. Arsenobetaine was the dominant form in the crustaceans and fish. The concentration of arsenic in the hot spring water was 750 μg/L and in the river water 17 μg/L. The highest level of arsenic in the biological samples was determined in the water bug and the green macroalga, 18,000 μg/kg and 18,000 μg/kg, dry weight, respectively. In the crustacean and fish muscle tissues, concentrations were 2600 ± 90 μg/kg and 150–2100 μg/kg, dry weight, respectively.

Naturally occurring dissolution from soil caused elevated levels of arsenic in the groundwater in the southern Fukuoka Prefecture, as reported by Kondo et al. (1999). Of the investigated 67 samples, in 29 the arsenic levels exceeded the established standard for drinking water, e.g. 0.01 mg/L, with the maximum determined concentration of 0.293 mg/L.

Seafood and rice are among the most investigated dietary products for their arsenic content. Hirata et al. (2011) analysed fish and prawn from Lake Biwa, a source of drinking water for the Kinki area in central Japan. In 2007 a mass mortality event of the endemic fish Isaza (*Gymnogobius isaza*) and the lake prawn (*Palaemon paucidens*) occurred in the lake. The authors found higher mean levels of arsenic and manganese, in the affected biological samples compared to fresh samples from the lake.

Rice, an important part of daily diet in Japan, is especially efficient in assimilation of arsenic. Signes-Pastor et al. (2009) studied total arsenic and arsenic speciation in rice and rice products (misos, syrups, amazake, barley, rice, and millet). The results showed higher content of inorganic arsenic in the investigated rice products, compared to barley and millet. Most of the total arsenic in rice products was inorganic (63–83%). The authors calculated that consumption of these products on regular basis could reach up to 23% of the provisional tolerable daily intake of arsenic, set by the WHO. Sun et al. (2009) reported concentrations of arsenic in the range from 0.14 to 0.28 mg/kg in 40 of the investigated rice products. Liquid rice products (oil, vinegar) and rice milk had the lowest concentration of As, even though higher than the water standard of 0.01 mg/L. However, the authors highlight that while the vinegar and oil contribute small part of the dietary intake, rice milk, as a beverage, could have a more significant contribution to the dietary intake of arsenic. Solid rice products contained higher levels of arsenic, and the highest were determined in the soil rice crackers. Oguri et al. (2014) found inorganic arsenic in 9 out of 19 food composites, prepared from products from the Shizuoka city in Japan. Source of the highest daily intake were cereals (13 μg/person/day) and algae (5.7 μg/person/day). Rice and hijiki were found to contribute most to the total daily inorganic arsenic intake by Japanese population, which is estimated to be 21 μg/person/day.

A group of arsenolipids was also found to exhibit negative effects on the human health. Their level in 17 food composites prepared from 152 food items from the Shizuoka city marketplace was investigated by Amin et al. (2018). The study confirmed marine food (algae, fish, and shellfish) as a predominant source of arsenolipids. Yorifuji et al. (2017) investigated health issues of survivors of arsenic poisoning by contaminated milk powder in infancy. The authors found decreased height in the exposed individuals as well as higher mean serum concentration of alkaline phosphatase (ALP).

7.7 Polynesia

Speciation of arsenic compounds in marine fish and shellfish from the two islands of the US territory of American Samoa, Tutuila and Ofu (South Pacific), showed only a minor fraction of inorganic arsenic, as found by Peshut et al. (2008). The concentrations of the total arsenic ranged from 0.235 to 98.2 μg/g, while the inorganic As was below detection limit in majority of samples and, if detectable, ranged from 0.0096 to 0.244 μg/g. The inorganic arsenic comprised less than 0.5% of the total arsenic, except for the few samples of molluscs in which inorganic arsenic ranged from 1% to 5%. In addition, Peshut et al. (2008) found no indications of biomagnification or bioaccumulation trends of arsenic in the investigated biota samples. The authors concluded that total arsenic is not a good indication of the possible arsenic toxicity for humans, highlighting the necessity of speciation analysis in these assessments.

8 Conclusion

Local geology and decades of mining activity, combined with increasing population density and increased water requirements, are the main reasons for the rise in arsenic concentration in surface water as well as groundwater, not only in Australia but also in Russian Federation and some African countries. Regardless of the origin of arsenic in drinking water, arsenic-contaminated water is usually the biggest contributor to total arsenic exposure in population of these regions. Additionally, rice as an agricultural crop that may contain elevated arsenic concentrations when grown on soils enriched with this element presents a staple food for a large part of the population in most of the developing countries in sub-Saharan Africa as well as in many Asian countries. Although the scarcity of data precludes a broader image on As contamination of drinking water and food products in the mentioned regions, reported figures emphasize the necessity of further research. In European countries, several regional hotspots of As contamination are mostly related to geogenic sources associated with bedrock lithology. Most notable is the case of the Pannonian Basin (Hungary, Serbia, and Romania), where more than 600,000 residents are at risk of drinking water containing high As concentrations. Other regions threatened by waterborne As include Czech Republic, Croatia, Finland, Greece, Italy, Spain, and Turkey. European research has indicated that there are large natural variations in As distribution in the environment and that geochemical maps at a variety of scales should be provided in the near future.

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