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Heavy metals in drinking water: Occurrences, implications, and future needs in developing countries

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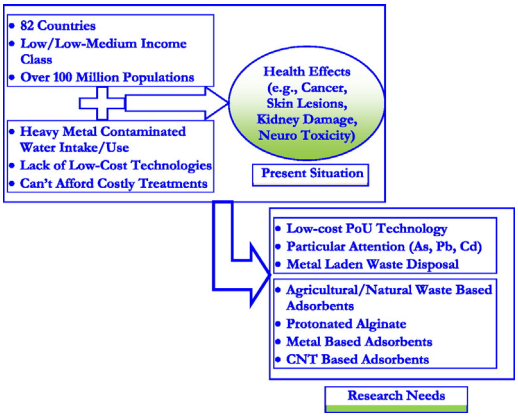
HIGHLIGHTS

- Co-exposure to multiple heavy metals in drinking water needs better understanding
- Low-cost technologies for arsenic removal needs urgent attention
- Protonated alginate needs further research for drinking water applications
- Community level and PoU devices need improvement and cost reduction
- Developing countries are most affected by heavy metals in drinking water

GRAPHICAL ABSTRACT



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ABSTRACT

Heavy metals in drinking water pose a threat to human health. Populations are exposed to heavy metals primarily through water consumption, but few heavy metals can bioaccumulate in the human body (e.g., in lipids and the gastrointestinal system) and may induce cancer and other risks. To date, few thousand publications have reported various aspects of heavy metals in drinking water, including the types and quantities of metals in drinking water, their sources, factors affecting their concentrations at exposure points, human exposure, potential risks, and their removal from drinking water. Many developing countries are faced with the challenge of reducing human exposure to heavy metals, mainly due to their limited economic capacities to use advanced technologies for heavy metal removal. This paper aims to review the state of research on heavy metals in drinking water in developing countries; understand their types and variability, sources, exposure, possible health effects, and removal; and analyze the factors contributing to heavy metals in drinking water. This study identifies the current challenges in developing countries, and future research needs to reduce the levels of heavy metals in drinking water.

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Contents

1. Introduction

Supply of safe drinking water is crucial to human life, and safe drinking water should not impose a significant risk to humans (WHO, 2011). Although few heavy metals are essential for human health, an excess amount of these metals can have negative effects (USEPA, 2015). The heavy metals are released into the environment through natural process and anthropogenic activities. The industrial processes generate wastes, which are mostly discharged into the environment. Industrial activities, especially electroplating, metal smelting and chemical industries, and manufacturing processes are few sources of anthropogenic heavy metals in water (He et al., 2008). Poorly treated domestic, industrial, and agricultural wastewater contains high concentrations of metals, which are often discharged into the environment in many developing countries (Gupta, 2008). Liu et al. (2011, 2012) identified industrial effluents and background sources as the main sources of heavy metals in the Hongfeng Lake sediments in China. Heavy metals can also enter into the environment through a bio-geo process (Szefer et al., 1999). The concentrations of arsenic in groundwater in 50 districts of Bangladesh were much higher than 50 μg/L, the limit established for Bangladesh (Uddin and Huda, 2011; Jiang et al., 2013). The floodplain and deltaic region were contaminated with arsenic (Chakraborti et al., 2010), and the main source was subsurface contamination (Karim, 2000). Heavy metals can be accumulated in sediments below the water columns and act as a source of metals through resuspension (Chowdhury et al., 2004; Wu et al., 2014; Wang et al., 2015). Few heavy metals, such as lead and mercury, can also enter the atmosphere due to traffic pollution and industrial activities, which can be deposited in soils around the reservoir and then enter the water along with the surface runoff (Wang et al., 2015). It is important to note that arsenic (As) is a metalloid and we will use the term "heavy metal" in this study.

The sources of drinking water (e.g., surface water, groundwater, and seawater) are likely to be polluted by heavy metals (Bryan and Langston, 1992). Leaching of metals from water distribution system (WDS) can contaminate drinking water (Al-Saleh and Al-Doush, 1998; Alabdula'aly and Khan, 2009). Although metals are removed during desalination of seawater, desalinated drinking water might contain various metals, possibly due to treatment and stabilization, blending with treated groundwater and leaching of metals from pipes of the WDS (Guo, 1997; Alabdula'aly and Khan, 2009). Heavy metal pollution in drinking water and the associated effects have been of increasing interest (Volety, 2008; Karavoltsos et al., 2008; Delpla et al., 2009; Liu et al., 2009, 2011; Montuori et al., 2013; Mandour and Azab, 2011; Shanbehzadeh et al., 2014). A number of heavy metals can induce cancer (e.g., inorganic arsenic [As]) and/or non-cancer (e.g., mercury [Hg]) risks in humans (USEPA, 2014, 2015). Studies have reported various chronic and sub-chronic effects from exposure to heavy metals. As in drinking water has affected >40 million people in Bangladesh (Karim, 2000; Chakraborti et al., 2010; Uddin and Huda, 2011). The Jinzu River water was found to be contaminated with cadmium from the Kamioka Zinc Mine in Japan, leading to kidney problems among the population (Yoshida et al., 1999). Few other studies have noted various health problems due to heavy metals in drinking water (Jaishankar et al., 2014; Zhang et al., 2014; Colak et al., 2015). To minimize the effects of heavy metals, regulatory agencies have proposed the maximum allowable limits in drinking water (e.g., WHO, 2011; USEPA, 2015).

Past studies have reported heavy metals in drinking water, including their types and quantities, factors affecting metal concentrations, sources, human exposure, risk, and removal. Despite significant progress, research is needed to ensure safe drinking water. Of particular concern, many low- and medium-income countries are faced with the challenge of reducing few heavy metals below the proposed limits, possibly due to their limited economic capacities (World Bank, 2016). Small and rural communities and individuals often consume water with a higher level of heavy metals than the guideline values (WHO, 2011). To date, the removal of all heavy metals from drinking water with a comprehensive technique has not been reported. Further, populations are exposed to drinking water from taps inside the building, where the metal concentrations increase due to stagnation of water in the WDS, cooler, hot water tanks (HWTs), and plumbing pipes (PP) inside the building.

In this paper, the research to date was divided into four major categories focusing on the occurrences and sources of heavy metals, human exposure and risks, removal, and critical gaps. The review identified the state of research to date, and summarized the findings of past research, limitations, and future research directions. The problems faced by developing countries were given particular attention. Finally, research strategies were identified to protect human health in developing countries.

2. Heavy metals in drinking water and their sources

Many heavy metals were reported in drinking water (Table 1). Drinking water is mainly produced from surface water, groundwater, and desalinating seawater, with desalination satisfying a significant fraction of the drinking water demand in water-scarce regions (Kim et al., 2015). Bottled water, spring water, and harvested rainwater are also used as drinking water in these regions (Sullivan, 2011). The sources of water, treatment, finished water quality, length and materials of the pipes of WDS, and PP may significantly contribute to the concentrations of heavy metals in drinking water.

In Sonora, Mexico, drinking water from wells and storage tanks were found to have elevated levels of As, Cd, Cu, Hg, and Pb. Approximately 43% of samples exceeded the action level for Pb (e.g., 15 μg/L) and 8.9% samples exceeded the guideline value of As of 10 μg/L (Wyatt et al., 1998; WHO, 2011; USEPA, 2015). The industrial, mining, and agricultural activities in the surrounding areas polluted the drinking water sources (Wyatt et al., 1998). In another study, the steel, plastic, and battery industries were found to be the major contributors of heavy metals in drinking water (Mandour and Azab, 2011). The concentrations of Cd, Co, Cu, and Pb in drinking water wells in 10 cities of Saudi Arabia were in the ranges 2–10, 5–30, 10–180, and 1–5 μg/L, respectively, which were attributed to the Gulf War and the Kuwaiti oil fires (Hashem, 1993). Tamasi and Cini (2004) reported higher levels of Fe, Cu, Zn, and Pb at the dead end of WDS than in the origin (e.g. treatment plants) in

Table 1
Sources of heavy metal contamination in drinking water and drinking water sources.

Metals	Medium	Location	Sources of contamination	References
Cu, Fe, Zn	Drinking water from the tap in multistory buildings	Dhahran, Saudi Arabia	Corrosion of pipe materials and coatings during water transportation and stagnation in plumbing systems	Alam and Sadiq (1989)
Cd, Co, Cu, Pb	Groundwater sources for drinking water	Ten cities of Saudi Arabia	Gulf War and burning of Kuwaiti oil wells	Hashem (1993)
As, Cd, Hg, Se	Desalinated product water from three plants	Jubail, Al-Khobar, and Al-Khafji, Saudi Arabia	Stabilization and pH adjustment of desalinated water and carryover of metals from source water	Kutty et al. (1995)
Pb, Cu, Cd, As, Hg	Drinking water wells and storage tanks	29 cities in the state of Sonora, Mexico	Industrial, mining, and agricultural activities near water sources	Wyatt et al. (1998)
Cd, Cu, Fe, Hg, Mn, Ni	Household drinking water	Riyadh, Saudi Arabia	Corrosion of metal pipes, water tanks, and plumbing systems.	Al-Saleh and Al-Doush (1998)
Hg	Bottled drinking water	Riyadh, Saudi Arabia	Industrial effluents, packaging activities of bottled water	Al-Saleh and Al-Doush (1998)
Cd, Be, Hg	Harvested rain water	Cairo and Giza, Egypt	Air pollution due to industrial activities	Saleh et al. (2001)
Cu, Cr, Fe, Mn, Ni	Surface water sources for drinking water	Northern Greece	Soil leaching	Simeonov et al. (2003)
Cd, Pb, Zn	Surface water sources for drinking water	Northern Greece	Heavy metal activities and industrial effluents	Simeonov et al. (2003)
As, Cu, Fe, Pb, Zn	Drinking water	Mount Amiata and downtown Siena, Italy	Leaching from water distribution pipes	Tamasi and Cini (2004)
Cd, Cu, Pb, Zn, Cr	Surface water source for drinking water	Dipsiz stream in Yatagan Basin, Turkey.	Coal-fired power plant	Demirak et al. (2006)
As, Mn	Groundwater (used as drinking water)	Southern Vietnam and neighboring Cambodia	Geogenic activities, and caused by natural anoxic conditions in the aquifers	Buschmann et al. (2008)
Cd, Cr, Cu, Ni, Pb, Zn	Surface water sources for drinking water	Ranipet industrial area, India	Industrial wastewater effluents and/or dumping of industrial waste	Gowd and Govil (2008)
Cd, Cu, Cr, Ni, Pb	Drinking water from WDS and tap	Several islands from nine regions in Greece	Anthropogenic activities, geogenic processes, seawater intrusion	Karavoltsos et al. (2008)
As, Cd, Pb, Se, Zn	Drinking water from tap	Dakhlia, Egypt	Corrosion of plumbing systems	El-Harouny et al. (2008)
Al, Cr, Cu, Fe, Mn, Ni, Pb, Zn	Drinking water stored in coolers	Riyadh, Saudi Arabia	Exposure to sunlight, absence of filtration device at the inlet, and aging of coolers	Alabdula'aly and Khan (2009)
As	Surface and groundwater sources of drinking water	Patancheru industrial town, Hyderabad, India	Paint, pharmaceutical, fertilizer, and pesticide industries; agricultural activities; As migration from surface to groundwater	Krishna et al. (2009)
Fe, Ni, Pb, Zn	Surface and groundwater sources of drinking water	Patancheru industrial town, Hyderabad, India	Industrial activities	Krishna et al. (2009)
Co, Cr	Surface and groundwater sources of drinking water	Patancheru industrial town, Hyderabad, India	Geogenic processes (natural concentration in granite terrain)	Krishna et al. (2009)
Ba, Sr	Surface and groundwater sources of drinking water	Patancheru industrial town, Hyderabad, India	Anthropogenic (e.g., pharmaceutical industries) and geogenic processes	Krishna et al. (2009)
Pb, Cd, Ni, Cu, Cr	Surface water sources from Buriganga River	Dhaka, Bangladesh	Industrial effluents, pesticides, domestic sewage, and tannery industry effluents	Ahmad et al. (2010)
As	Drinking water from shallow tube wells (<300-m depth)	Floodplain and deltaic and coastal regions of 64 districts in Bangladesh	Subsurface contamination of Holocene floodplain deposits in 59 out of 64 districts	Chakraborti et al. (2010)
Ni, Cr, Pb, Cu, Hg	Surface water sources in Upper Lake	Bhopal, India	Industrial activities, geogenic processes	Virha et al. (2011)
Cd, Mn, Pb	Groundwater used as drinking water (tube, deep tube, and ring wells)	Assam, India	Geogenic contamination	Chakrabarty and Sharma (2011)
Cd, Pb	Drinking water from WDS and tap	Dakahlia, Egypt	Steel, plastic, and battery industries and household plumbing systems	Mandour and Azab (2011)
Cd, Cu, Pb	Groundwater as source of drinking water	Batlagundu, Tamil Nadu, India	Industrial and human activities related to urbanization	Thambavani and Mageswari (2013)
As, Cr, Se	Groundwater wells as sources of drinking water	Makkah, Saudi Arabia	Geological features surrounding Makkah	Khdary and Gassim (2014)
Pb	Rainwater harvesting systems	Southwest coastal areas of Bangladesh	Atmospheric dissolution	Islam et al. (2014)

Mount Amiata and downtown Siena, indicating the leaching of metals from the water distribution pipes into water.

El-Harouny et al. (2008) reported the corrosion of PP as the major source of contamination of drinking water in Dakhlia, Egypt. Alabdula'aly and Khan (2009) reported that stagnation of water in coolers for several hours increased the concentrations of Al, Cr, Cu, Fe, Pb, Mn, Ni, and Zn significantly. The concentrations of As and Mn in groundwater-sourced drinking water in the Mekong Delta floodplains of Southern Vietnam and neighboring Cambodia ranged from 0.1 to 1340 μg/L and from 1.0 to 34.0 mg/L, respectively. In this study, approximately 37% of well-sourced drinking water exceeded the regulatory limit of As (Buschmann et al., 2008; WHO, 2011). The contamination was geogenic and was caused by natural anoxic conditions in the aquifers (Buschmann et al., 2008). In Greece, Karavoltsos et al. (2008) demonstrated heavy metal contamination of drinking water due to anthropogenic and natural contamination of groundwater and seawater intrusion into the aquifers, whereas seawater intrusion is a common problem in most coastal regions across the world.

In Lagos, Nigeria, Momodu and Anyakora (2010) reported 32.7% and 36.7% of drinking water well samples with higher Cd and Pb levels, respectively, than the guideline values (WHO, 2011). In Assam, India, geogenic contamination has resulted in higher levels of Cd, Mn, and

Pb (averages: 25, 17, and 27 μg/L, respectively) in drinking water from shallow and deep tube wells and ring wells (Chakrabarty and Sharma, 2011). Drinking water near the tea gardens of Sonitpur district, Assam, showed As levels up to 90 μg/L (Dutta, 2013). The concentrations of As, Ni, and U in drinking water in the territory of Kosovo exceeded the World Health Organization (WHO) guidelines in 3.1%, 2.1%, and 7.3% of the total samples, respectively (Berisha and Goessler, 2013). Drinking water from bored and pumped drilled wells often had higher concentrations of As, Ni, and U, possibly due to the geogenic contamination (Berisha and Goessler, 2013). The average concentrations of Cr, Mn, Ni, Pb, and Zn in drinking water in rural areas of Bhongiri, India, were 39.9, 147.5, 13.4, 14.4, and 1286.1 μg/L, respectively, in which significant fractions of samples exceeded the WHO guidelines of 0.05, 0.07, and 0.01 mg/L for Cr, Ni, and Pb, respectively (Chennaiaha et al., 2014; WHO, 2011). Chakraborti et al. (2010) summarized 14 years of data on As contamination of groundwater-sourced drinking water using 52,202 samples from 64 districts in Bangladesh. Approximately 27.2% and 42.1% of the samples had As above 50 and 10 μg/L, respectively, whereas 7.5% of the total samples contained As above 300 μg/L. The groundwater of 50 districts had As above the Bangladesh standard (50 μg/L) and 59 districts had above the WHO guideline (10 μg/L) during the 14 years (1996-2009). The tube wells in the tableland and hill tracts were free from As, whereas the floodplain and deltaic region, including the coastal region, were highly contaminated with As. The main source of As was subsurface contamination of groundwater by the Holocene floodplain deposit (Karim, 2000; Chakraborti et al., 2010).

In water-scarce regions (e.g., Saudi Arabia and California), drinking water is often supplied by blending groundwater with desalinated seawater (MOWE, 2014; SWCC, 2014). However, this is not a viable option for low- and medium-income countries, and thus heavy metals in desalinated and blended water are not included here. Bottled water, which is used universally, can have higher levels of heavy metals. Sullivan (2011) reported lower levels of As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Pb, Se, V, and Zn than the Maximum Contaminant Levels (MCLs) as defined by the United States Environmental Protection Energy (USEPA) in six sources of bottled natural spring water in California. Saleh et al. (2001) reported higher concentrations of Na, Mg, Si, V, Cu, Fe, and Sb in five major brands of bottled water than in tap and rainwater from Cairo and Giza in Egypt (Saleh et al., 2001). Al-Saleh and Al-Doush (1998) reported higher concentrations of Hg in 21 brands of retail bottled water in Riyadh, Saudi Arabia, with packaging activities identified to be the possible sources of Hg pollution. El-Harouny et al. (2008) reported relatively higher levels of Pb, Cd, and As and lower levels of Hg in tap water than in three commercial brands of bottled water in Egypt. Further, rainwater was reported to contain higher levels of Cd, Be, and Hg than tap and bottled water, indicating possible air pollution due to industrial activities and/or traffic pollution (Saleh et al., 2001).

Drinking water can contain heavy metals originating from contaminated source water. In 22 samples, high concentrations of As (average: 29.2 μg/L; range: 0–116.5 μg/L) were reported for the surface water of Patancheru industrial town, Hyderabad, India, which were linked to the paint, pharmaceutical, fertilizer, and pesticide industries (Krishna et al., 2009). Groundwater from the same location (31 samples) showed much higher concentrations of As (average: 146.5 μg/L; range: 2.9–1257 μg/L), possibly due to the migration of As from surface water and geogenic contamination, which was caused by alluvium deposits of 5–10-m thickness over the granitic terrain in that area (Krishna et al., 2009). In addition, the levels of Sr, Ba, Co, Ni, Cr, As, Mn, Pb, and Zn in the surface water and groundwater were relatively higher (Krishna et al., 2009). In another study, the rivers around Dhaka, Bangladesh, showed high levels of Pb, Cd, Ni, Cu, and Cr, which were attributed to industrial effluents, extensive use of pesticides, waterway transportation, discharge of untreated domestic sewage, and the tannery industry effluents (Ahmad et al., 2010). Near the effluent discharge locations, these metals were found in the ranges of 58.2–72.5, 7.1–12.3, 7.2–10.3, 107.4–201.3, and 489.3–645.3 μg/L, respectively (Ahmad et al., 2010). Gowd and Govil (2008) reported average concentrations of Cd, Cr, Cu, Ni, Pb, and Zn in surface water from the Ranipet industrial area, India, of 51.1, 247.2, 95.5, 36.7, 467.8, and 3760.4 μg/L, respectively, which were attributed to the industrial wastewater effluents and/or dumping of industrial waste (Gowd and Govil, 2008). Virha et al. (2011) demonstrated the anthropogenic contamination of the Upper Lake, which was the major source of drinking water in Bhopal, India. Khdary and Gassim (2014) reported Cr, As, and Se in the wells of Makkah, Saudi Arabia, with 42% of samples exceeding the guideline value of 10 μg/L for As.

The past studies reported many sources of heavy metal contamination in drinking water, some of which are presented in Table 1. As, Cd, Cu, Ni, Pb, and Zn were frequently detected in high concentrations in many regions (Table 1). Industrial activities, pesticides, domestic wastewater, and tannery effluents were the major sources of Cd, Cu, Ni, Pb, and Cr in drinking water. The major sources of As were geogenic, primarily due to subsurface contamination from floodplain deposits and natural anoxic conditions in the aquifers; certain types of industries and agricultural activities were also the sources of As in drinking water (Table 1). Corrosion of pipes, pipe coatings, coolers, and water tanks was also a significant source of Cu, Pb, Ni, Cr, Fe, and Zn in drinking water. Hg in drinking water mainly originated from industrial activities, packaging for bottled water, corrosion of pipe materials and coatings, amalgam in teeth, and powder laundry detergents, and a few other sources of Hg include agriculture, wastewater, mining, and incineration. The chemicals used in water treatment plants were major contributors of Ni in drinking water (Table 1).

3. Human exposure and risks

Thirty-five metals pose a threat to human health because of residential and/or occupational exposure, 23 of which are heavy metals (Mosby et al., 1996). Studies have reported various effects of heavy metals in drinking water (ATSDR, 2015; USEPA, 2015). According to the International Agency for Research on Cancer (IARC), inorganic As and Cd are classified as human carcinogens (IARC, 2016). As is related to cancer risk and skin damage, whereas Cd is linked to kidney damage and cancer. Other effects such as heart diseases and blood cholesterol from Sb, anemia from Pb, kidney and liver damage from Hg, and gastrointestinal disorder from Cu were also reported (ATSDR, 2015; USEPA, 2015). Among the heavy metals, As, Cd, Pb, Cr, Cu, Hg, and Ni are of major concern, mainly due to their presence at relatively high concentrations in drinking water and their effects on human health (ATSDR, 2015). Table 2 summarizes some effects among populations exposed to heavy metals.

Among the heavy metals, As, Cd, and Pb were extensively studied for their public health effects (Fu et al., 2013; ATSDR, 2015; USEPA, 2015). Smith et al. (1992) reported that drinking 1 L/day water with As of 50 μg/L over one's lifetime could lead to cancer of the liver, lung, kidney, or bladder in 13 per 1000 persons. Another study reported an increased incidence of skin lesions from an As dose of 0.0012 mg/kg/day through drinking water (Ahsan et al., 2006). Arain et al. (2009) reported moderate correlation between As in drinking water and respiratory disorders. As was reported to be detrimental to the central nervous system and cognitive development in children (Rosado et al., 2007). It was also found to accumulate in fingernails and hair (Choong et al., 2007). Subchronic effects, such as the risk of stillbirths, were increased by six fold in women who consumed drinking water with As ≥200 μg/L during pregnancy (von-Ehrenstein et al., 2006). As in drinking water is one of the major issues in >30 countries (Chowdhury et al., 2000). In Bangladesh, approximately 6.8 million people were exposed to groundwater contaminated with As above 300 μg/L (Chakraborti et al., 2010). Arsenical skin lesions were common among the populations in Bangladesh (Chakraborti et al., 2004). In Bangladesh, it could cause 200,000–270,000 deaths from cancer (Smith et al., 2000). Few other known As-affected regions include West Bengal, Hyderabad, and Assam of India (von-Ehrenstein et al., 2006; Krishna et al., 2009); Kurdistan of Iran

Table 2
Metal contamination and effects on public health.

Cause(s)	Observed or effects	References
Cadmium pollution in the Jinzu–Takahara River (≅10 μg/L) and air by the Kamioka mine industry in Japan	Osteomalacia, softening of the bones due to renal tubular dysfunction (Itai-Itai disease). The mining company had to provide compensation based on the verdict of Japanese court.	Yoshida et al. (1999)
As contamination of groundwater (drinking water) in 50 out of a total of 64 districts in Bangladesh	Approximately 200,000–270,000 deaths from cancer linked to As-contaminated drinking water consumption in Bangladesh were estimated	Smith et al. (2000)
Mean U level in drinking water was 131 μg/L in the range of 0–1920 μg/L. 30% samples had U >100 μg/L. Median daily intake was 39 μg (7–224).	Nephrotoxic effects were reported from U in drinking water from drilled wells in Finland. U intake was related to increased fractional excretion of Ca. In urine: 13 ng/mmol creatinine (2–75) was observed. Increased systolic and diastolic blood pressures were also observed.	Kurttio et al. (2002)
Consumption of As-contaminated drinking water. As levels were 0.14, 37.9, 171.6, and 611.9 μg/L, respectively, in four town/villages in Eastern Croatia	Mean As levels in hair were 0.07, 0.26, 1.74, and 4.31 μg/g, respectively. About 3% of the Croatian population may be affected due to As exposure.	Cavar et al. (2005)
Exposure to As-contaminated drinking water (≥200 μg/L) during pregnancy	Sixfold increased risk of stillbirth in the studied area of West Bengal, India	Von-Ehrenstein et al. (2006)
Consumption of As-contaminated groundwater (as drinking water) without treatment	As poisoning could put two million people in the Mekong Delta (Southern Vietnam and bordering Cambodia) at risk of cancer and skin diseases	Buschmann et al. (2008)
Higher levels of Cd, Cr, Cu, Ni, Pb, and Zn in surface water. Pollution was caused by the industrial effluents from Ranipet, India	People were adversely affected and suffered from asthma, chromium ulcers, and skin diseases. The area had approximately 240 tanneries, ceramic industries, refractory boiler auxiliaries, and chromium plant industries.	Gowd and Govil (2008)
Consumption of As-contaminated drinking water in eight villages in Kurdistan, Iran. Average levels of As were 0–460 μg/L in these villages.	Lesions on skin and liver, hyperkeratosis or hyperpigmentation, and respiratory complications.	Mosaferi et al. (2008)
Exposure to low levels As (average: 1.2 μg/L; range: 0.05–25.3 μg/L) in drinking water in Denmark	Exposure to low doses of As might reduce the risk of skin cancer.	Baastrup et al. (2008)
As-polluted drinking water (avg.: 97.5; range: 35.2–158 μg/L) consumption in southern Sindh, Pakistan.	As-induced skin lesions, higher levels of As in hair and blood sample	Arain et al. (2009)
Approximately 36.6 and 22.7 million people in Bangladesh could be exposed to drinking water with As above 10 and 50 μg/l, respectively. Up to 6.8 million could be exposed to water with As above 300 μg/l.	Skin diseases including melanosis, leukomelanosis, keratosis, hyperkeratosis, dorsum, non-pitting edema, gangrene, and skin cancer have been identified among several million people. Approximately 6.8 million people could have arsenical skin lesions.	Chakraborti et al. (2010)
Consumption of Cd- and Pb-polluted water. Cd-contaminated phosphate manures and Pb-containing pesticides were the main sources of pollution.	Chronic renal failure among the population in a village in Buldana district, Maharashtra, India.	Bawaskar et al. (2010)
Higher levels of Cd, Cr, Fe, and Pb in tap and bottled drinking water. The study was carried out in Shebin El-Kom City, Menoufiya, Egypt.	Blood creatinine, urea, and hemoglobin levels were positively correlated with one or more of Cd, Cr, Fe, and Pb. Effects on kidney function and hemoglobin levels were noted.	Badr et al. (2011)
Nonoccupational exposure to Cd, Hg, and Pb from dietary and environmental sources and cosmetics (e.g., dental amalgam fillings, skin-lightening cream).	At childbirth, many mothers had Cd, Hg, and Pb in the cord, maternal blood, and placenta. The study was performed at the King Khalid Hospital at Al-Kharj, Saudi Arabia, between 2005 and 2006	Al-Saleh et al. (2011)
Consumption of Co, Cr, Cu, Fe, Mn, Ni, and Pb through drinking water.	Hazard quotients for ingestion and dermal routes were in the ranges of 0.011–0.14 and 0.000085–0.098, respectively, in Karachi, Pakistan.	Karim (2011)
Contamination of eight heavy metals (As, Se, Hg, Cd, Ag, Mn, Cr, and Pb) in drinking water. As levels in drinking water were very high (42–1500 μg/L).	Strong linear relationship between As intake and multi-chronic arsenical poisoning in 530 villages in Kurdistan, Iran. About 30.7% of participants had chronic arsenical poisoning. Mee's lines, keratosis, and pigment disorders were present in 86.1%, 77.2%, and 67.8%, respectively.	Alasvand et al. (2012)
Contamination of water in the Zhalong Wetland, China, with Cu, Cd, Cr, As, Zn, and Ni	Risks due to Cu, Cd, Cr, As, Zn, and Ni in the Zhalong Wetland, China, were lower than the maximum allowable levels.	Zhang et al. (2014)
Contamination of shallow groundwater wells (drinking water) by As, Cd, Cr, Cu, Hg, Ni, Zn, and Pb due to agricultural activities	Hazard indices exceeded acceptable limits in 58% of the tested wells in Hua-ruea subdistrict, Ubon Ratchathani, Thailand. Cancer risks were higher than the acceptable level in one well.	Wongsasuluk et al. (2014)
Heavy metal contamination of drinking water	Be, Ni, and Mo were negatively and Sb was positively associated with cancer incidence in the residential areas of the Black Sea region, Turkey.	Colak et al. (2015)

(Mosaferi et al., 2008; Alasvand et al., 2012); Southern Vietnam and bordering Cambodia of the Mekong Delta floodplain (Buschmann et al., 2008); Southern Sindh of Pakistan (Arain et al., 2009); Inner Mongolia of China (Wade et al., 2009; Xia et al., 2009); and Eastern Croatia (Cavar et al., 2005). Northern Afghanistan, Northern Mali and Zambia in Africa (Amini et al., 2008); Thailand and Taiwan in Asia (Ning, 2002); and Mexico, El Salvador, Peru, and Nicaragua in the Americas (Bundschuh et al., 2006) have also reported higher concentrations of As in drinking water. In Mexico, the largest number of the dispersed populations was exposed to drinking water with un-healthy levels of As, and with arsenicosis as a public health issue (Bundschuh et al., 2006; Armienta and Segovia, 2008).

Next to As, Pb was most investigated in the context of its public health effects. Pb can affect the central nervous, renal, hematopoietic, cardiovascular, gastrointestinal, musculoskeletal, endocrinological, reproductive, neurological, developmental, and immunological systems (ATSDR, 2015). Exposure to low levels of Pb can affect neurodevelopment. During brain development, the level of lead in the blood (PbB) interferes with the trimming and pruning of synapses, migration of neurons, and neuron/glia interactions. Alterations of any of these processes may result in failure to establish appropriate connections between structures and eventually in permanently altered functions (ATSDR, 2015). However, to date, no threshold has been

established for such effects (ATSDR, 2015). In a study on 4853 children of ages 6–16 years, Lanphear et al. (2000) reported a decrease in arithmetic and reading skills for a PbB<5 μg/dL. Canfield et al. (2003) observed a decline of 7.4 points in the intelligence quotient (IQ) among 172 children of age 5 years with a PbB increase of 1–10 μg/dL. Similar findings were reported by Chen et al. (2005). Chiodo et al. (2004) observed decreased attention, executive function, visual motor integration, and motor skills among 237 children of age 7.5 years at a mean PbB of 5.4 μg/dL, and they observed associations at 3.0 μg/dL. Wright et al. (2003) analyzed the cognitive test performance of 736 adults; they reported impaired performance for average PbB of 4.5 μg/dL and patella Pb of 29.5 ppm. Hu et al. (2006) demonstrated the effects of PbB in a study on 146 children (12 and 24 months old). Occupational exposure to Pb led to an increase in the systolic blood pressure among 496 adults at an average age of 56 years (Glenn et al., 2003). Hu et al. (2006) observed an increase in hypertension from tibia Pb content of 8–37 ppm . Although significant fractions of PbB can be attributed to drinking water (Dudi et al., 2005; Triantafyllidou et al., 2007), other sources such as Pb paint, dust, and leaded gas can be the important sources of Pb (ATSDR, 2015).

The third most frequently reported heavy metal in drinking water is Cd, and it has been pointed as a public health concern (NTP, 2015; Fernández-Luqueño et al., 2013; ATSDR, 2015; USEPA, 2015). Cd-contaminated drinking water was linked to chronic renal failure (Bawaskar et al., 2010; ATSDR, 2015). Gobe and Crane (2010) reported kidney failure due to long-term exposure to Cd. Chronic exposure to Cd could lead to anemia, anosmia (loss of sense of smell), cardiovascular diseases, renal problems, osteoporosis, and hypertension (Yoshida et al., 1999; ATSDR, 2015). Cd can bind with cysteine, glutamate, histidine, and aspartate ligands and can cause iron deficiency (Castagnetto et al., 2002; Irfan et al., 2013). It can cause both acute and chronic intoxications (Chakraborty et al., 2013). It is highly toxic to the kidney, and it accumulates in the proximal tubular cells at higher concentrations (e.g., bio-accumulative). Jaishankar et al. (2014) reported that Cd can cause bone mineralization either through bone damage or by renal dysfunction. Upon a very long exposure time at lower concentrations, it could become deposited in the kidney, ultimately leading to kidney disease, fragile bones, and lung damage (Bernard, 2008). Experiments on animals have shown that 50% of Cd is absorbed in the lungs and less in the gastrointestinal tract (Jaishankar et al., 2014). Henson and Chedrese (2004) reported the association of Cd exposure during pregnancy with premature birth and reduced birth weights.

Few other heavy metals can also pose a threat to public health (ATSDR, 2015). Kurttio et al. (2002) reported the nephrotoxic effects and the increase in systolic and diastolic blood pressures due to U in drinking water in Finland. Gowd and Govil (2008) reported higher chromium levels in water as well as chromium ulcers among the population surrounding an industrial area in India. Another study in Greece with Cr concentrations of 8.3–51 μg/L in drinking water reported significantly higher standard mortality ratios for primary liver cancer, lung cancer, kidney cancer, and other genitourinary organs among women (Linos et al., 2011). Colak et al. (2015) reported a positive correlation between Sb and cancer incidence in the residential areas of the Black Sea region, Turkey. Exposure to Cu through contaminated water or other beverages resulted in nausea, vomiting, and/or abdominal pain (Araya et al., 2003). Drinking water was reported to be the primary source of excess Cu intake by humans (ATSDR, 2015). Exposure to elevated levels of Mn in drinking water during pregnancy might impair the intellectual development of children (Wasserman et al., 2006). In addition to As and Pb, Mn has also been identified as a developmental neurotoxicant (Grandjean and Landrigan, 2014). In Bangladesh, exposure to Mn in drinking water was found to reduce performance in mathematics among schoolchildren (Khan et al., 2012). A study in Quebec, Canada, demonstrated a strong correlation between Mn concentrations in hair and hyperactivity (Bouchard et al., 2007). In addition, associations were reported between Mn concentrations and reduced intellectual function, impaired motor skills, and reduced olfactory function (Grandjean and Landrigan, 2014).

To protect human health, regulatory agencies have recommended the maximum concentrations of few heavy metals (e.g., As, Cd, Cr, Hg, Pb, Se, and Sb) in drinking water (e.g., USEPA, 2015; WHO, 2011). The guideline values were determined based on the no-observed-adverse-effect limit (NOAEL), the lowest concentration that was reasonably and economically achievable with water treatment technologies, measurable lowest concentration, and the observed effects on human and/or animal health (WHO, 2011). Although the guideline values are considered to be health protective, co-exposure to multiple metals may need further attention. Co-exposure to multiple metals in drinking water might increase or decrease the level of toxicity. Past studies have demonstrated that the effects of As with respect to genotoxicity and metabolism could be aggravated due to co-presence with Sb (Gebel, 1997). McCarty et al. (2004) reported co-exposure of As and Sb in Bangladesh, which could have increased As toxicity. The co-presence of As (>10 μg/L), Ni (>1.0 μg/L), Cd (0.15 μg/L), Co (2.5 μg/L), and Cr (2.4 μg/L) was reported in the Mekong Delta floodplains (Buschmann et al., 2008). In the presence of one toxic metal, the second metal could exhibit the effect even at one-twentieth the WHO guideline value (Escher and Hermens, 2002). Further, humic substances in water might also increase As toxicity (Lamm and Kruse, 2005). These humic substances form complexes with inorganic As, which is released in the gastrointestinal tract, where it is absorbed (Tseng, 2005). By contrast, co-exposure of As and Se led to significantly low toxicity (Biswas et al., 1999). Further, Zn was also reported to reduce As toxicity, with the WHO recommending taking 15 mg of Zn daily to reduce the adverse health effects of As (Buschmann et al., 2008). It is essential to understand the synergistic or antagonistic health effects from co-exposure to multiple metals in drinking water. When multiple toxic metals are present in drinking water, the effects on human health might be observed at a level lower than the regulatory limits of these metals. Even at the low levels of the individual metals, the cumulative risks might exceed the allowable risk levels. Regulatory agencies should thus look into this issue to ensure better protection of human health. In addition, these agencies generally collect data from the WDS. Studies have shown that stagnation of water in the WDS, HWT and PP could significantly increase the concentrations of several metals. Populations are exposed to tap water in the house or multistory buildings where water can remain stagnant for long periods, thus increasing the metal concentrations. To establish the acceptable risk levels or NOAEL for a metal, an understanding of the effects of water stagnation in the plumbing premise (HWT and PP), and the co-exposure to multiple metals is necessary.

4. Removal of heavy metals from drinking water

Removal of heavy metals is an important step toward safe drinking water. Studies on heavy metal removal from drinking water have focused mainly on As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, and Zn (Fu et al., 2013; ATSDR, 2015). In the past, several hundred studies have reported various experiments to remove and/or sequestrate heavy metals from wastewater (Periasamy and Namasivayam, 1996; Kurniawan et al., 2006; Srivastava and Majumder, 2008; Kumar et al., 2011; Diale et al., 2011), groundwater (Hashim et al., 2011; Parimal, 2015), and drinking water (Upadhyayula et al., 2009; Yang et al., 2013; WHO, 2011; Parimal, 2015). Table 3 summarizes a few methods used to remove heavy metals from drinking water. Adsorption was found to be the method of choice with various adsorbents being available for this purpose (Fu et al., 2013). The other methods included chemical precipitation, physical separation, ion exchange, membrane filtration, membrane distillation, and hybrid methods (WHO, 2011; Parimal, 2015). Some of these methods (e.g., ion exchange, membrane filtration, etc.) are expensive and likely to be impractical for application in low- and medium-income countries (Fig. 1). In the following paragraphs, the discussion is limited

Table 3
Removal of heavy metals from drinking water.

Target metal removal	Materials and approach	Ref.
Removal of Cd, Co, Cu, Mn, Ni, and Pb from tap water containing high levels of Ca and Mg with a cation-exchange resin possessing a chelating iminodiacetic acid group. About 99% of metals were removed.	An 18-in.-diameter column was packed with 100 mL of Lewatit TP-207 (Bayer Co.) cation-exchange resin. The metals were introduced into tap water in the range of 0.8–1.2 mg/L. The water showed Ca, Mg, and bicarbonate levels of 80, 40, and 220 mg/L, respectively.	Korngold et al. (1996)
Removal of Cu from water and Cu plating industry wastewater. Adsorption capacities of PHC and commercial GAC were 65.6 and 3.6 mg/g, respectively. The contact time for maximum removal by PHC was 2.5 times less than that required by GAC.	Activated carbon prepared from peanut hull (PHC) was used for Cu adsorption from wastewater obtained from a Cu plating industry, Mettur, India. PHC was derived from agricultural waste by-products, and the cost was less than the commercial GAC.	Periasamy and Namasivayam (1996)
Removal of Cd, Cu, and Pb from water using metallurgical solid wastes as sorbents in Turkey. Red muds and fly ashes showed a high adsorption capacity for heavy metals in the order Cu > Pb > Cd (order of insolubility of metal hydroxides).	Low-cost bauxite red muds and coal fly ashes were used as sorbents. Metal solutions of 10,000 mg/L concentration were prepared using nitrate salts. Three groups of sorbents (washed and/or sieved, acid treated, and heat-treated) were prepared from red mud and fly ash. Batch sorption tests were performed using 1.0 g of sorbent in 50 mL of metal nitrate solution for 8 h at 25°C.	Apak et al. (1998)
Removal of Cd, Cr, Ni, Pb, and Zn from drinking water in Jordan. Pb and Zn were removed effectively, but the adsorption capacity for other metals was much less.	Processed solid residue of olive mill products (SROOMP) was used. The SROOMP passed through a sieve of 2 mm and retained at 1.18 mm was used. The cost of SROOMP was less than $50/ton, whereas that of commercial GAC was about $4500/ton.	Gharaibeh et al. (1998)
Removal of Cd from natural water used as drinking water source. Efficient elimination of Cd was found. Weakly basic ion exchangers may favor selective elimination of Cd from source waters.	Two commercially available weakly basic anion exchangers (Duolite A7 and Purolite A830) and one laboratory sample (Purolite A832) were used. The resins were pretreated with HCl and NaOH.	Zhao et al. (2002)
Removal of Cu, Pb, and Zn from drinking water using GAC in a point-of-use (PoU) water filter. Binary mixtures of acid-activated almond shell-based carbon with either steam-activated pecan shell- or walnut shell-based carbon removed nearly 100% of Pb, 90–95% of Cu, and 80–90% of Zn.	GAC was prepared from the shells of almond, English walnut, and pecan, which was used in a PoU water filter. The filter needed less carbon than four commercial brands in USA, and the production cost of a filter was less than US$ 1.0.	Ahmedna et al. (2004)
Removal of As and Hg from drinking water using an ion-exchange membrane bioreactor (IEMB). The IEMB process offered several advantages over other processes.	IEMB process was used where cation- and anion-exchange membranes were used for Hg and As, respectively. A strain of *P. putida* and sulfate-reducing bacteria converted Hg and As.	Oehmen et al. (2006)
Removal of Mn and Zn from drinking water using clinoptilolite (Clin) and clinoptilolite–iron oxide (Clin–Fe). Mn adsorptions of Clin and Clin–Fe were 7.69 and 27.12 mg/g, respectively, whereas the Zn adsorptions were 71.3 and 94.8 mg/g, respectively.	Natural clinoptilolite from Thrace, North Greece, was added to iron nitrate [$Fe(NO_3)_3$] solution under strongly basic conditions (5 M KOH) to develop Clin-Fe. Clin–Fe had a specific surface area of 151.0 m^2/g and was fully iron exchanged (Fe/Al = 1.23).	Dimirkou and Doula (2008)
Removal of Pb, Cu, and Cr from tap water using hybrid inorganic/organic adsorbents based on alumina and 4-aminoantipyre. The removal was in the range 93–97%.	Three hybrid inorganic/organic adsorbents (acidic, neutral and basic), prepared from alumina and 4-aminoantipyre, were used. The adsorbents were strongly resistive to acid leaching in pH range of 1–7 and thermally stable up to 350°C.	Mahmoud et al. (2010)
Removal of Cd, Pb, Fe, and Mn from drinking water using hybrid silica-based synthesized adsorbents. The mono-amino-functionalized silica was the best adsorbent.	Diverse amino-functionalized organic–inorganic hybrid adsorbents, developed through synthesis with silica, were used.	Xia et al. (2010)
Removal of As, Cr, and Pb from drinking water using the hierarchical porous magnetic Fe_2O_3@AlO(OH) superstructure nanomaterial. Adsorption was ultrafast, and removal efficiencies were very high.	The new composite nanomaterial composed of magnetic urchin-like α, γ-Fe_2O_3, and boehmite γ-AlO(OH) was used. The Fe_2O_3–AlO(OH) composite nanomaterial was synthesized by co-dispersing α, γ-Fe_2O_3, and $Al(NO_3)^3.9H_2O$.	Yang et al. (2013)
Removal of As, Cr, and Pb through adsorption and magnetic separation using tunable smart magnetic graphene (SMG). Approximately 99% removal of these metals was achieved.	The SMG, prepared using 50 mg of graphene oxide and 150 mg of ferrocene to a quartz tube containing 20 pieces of broken silicon wafers, which was then evacuated under vacuum for 30 min and irradiated inside a focus microwave oven, was used.	Gollavelli et al. (2013)

to low-cost techniques; thus, low-cost chemical precipitation and adsorbents are described in detail.

Synthetic activated carbon-based adsorbents were developed using natural materials (e.g., coconut shell, almond shell, fertilizer slurry, palm tree cobs, petroleum coke, pine saw dust, bituminous coal, lignite, fly ash, peanut shell, olive by-products, rice husk, seafood industry by-products, and tamarind wood), which were treated with different functional groups depending on the target metals (Table 3). Gharaibeh et al. (1998) used the processed solid residue of olive mill products (SROOMP), an agricultural by-product, to remove Cd, Cr, Ni, Pb, and Zn from drinking water. Pb and Zn were removed effectively. The cost of SROOMP was less than US$0.05/kg. Ahmedna et al. (2004) used binary mixtures of acid-activated almond shell-based carbon with steam-activated pecan shell or walnut shell-based carbon in the point-of-use (PoU) filter to remove Cu, Pb, and Zn from drinking water. The filters removed nearly 100% of Pb, 90–95% of Cu, and 80–90% of Zn. Moreover, the filters needed less carbon than the four commercial brands available in the USA, and the production cost of 1 kg of carbon was US$8.71. Assuming 110 g of carbon in each filter, the average cost of a filter was estimated to be less than US$ 1.0. Singh et al. (2008) prepared activated carbon from tamarind wood by chemical activation with sulfuric acid, which was found to remove almost 98% of Pb from water at initial Pb concentrations of 40 mg/L, with an adsorbent dose of 3.0 g/L at a pH of 6.5. Tamarind wood activated at pH 5.41 removed >89% of Cr from water (Sahu et al., 2009). Dimirkou and Doula (2008) demonstrated that clinoptilolite–iron oxide more efficiently removed Mn and Zn from drinking water than clinoptilolite alone. Santona et al. (2006) used red mud to remove Cd, Pb, and Zn from drinking water. Fine et al. (2005) applied activated carbon and peat to remove Cd and Cr, whereas Xenidis et al. (2010) used natural zeolites for the removal of Cd, Cr, Cu, and Ni. Industrial by-products and wastes were used for Cd and Pb removal (Mohan et al., 2007), whereas ferrous materials were used for Cu, Cr, Cd, Hg, and Pb removal from drinking water (Chowdhury and Yanful, 2010).

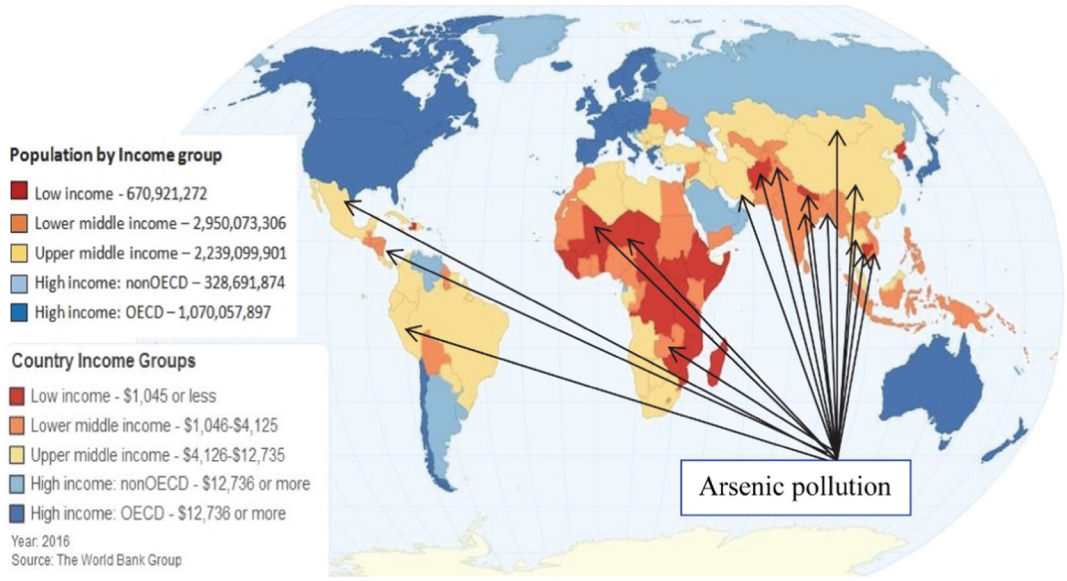

Fig. 1. Distribution of population according to income groups.

Lagoa and Rodrigues (2007) evaluated the performance of dry protonated calcium alginate beads for the biosorption of Pb. The maximum uptake capacity was estimated to be 339 mg/g, indicating that these beads were among the best biosorbents for the treatment and recovery of Pb from aqueous streams. Gomez et al. (2012) demonstrated the superior performance of calcium alginate to sodium alginate in removing Pb from aqueous solutions. More than 98% of Pb was removed by calcium alginate at an initial concentration of 0.1 mM. Salisu et al. (2015) applied sodium alginate-graft-poly (methyl methacrylate) in the form of calcium cross-linked beads to remove Pb from aqueous solutions. The maximum adsorption capacity was found to be 526 mg/g for concentrations ranging from 200 to 1000 mg/L. Most of the alginate sorbents were applied for higher initial concentrations of Pb than the usual concentrations in drinking water. Further, information on the cost of using different alginates is limited. Their performance at lower concentrations of Pb and the overall costs require further study.

Kurniawan et al. (2006) reported several agricultural waste-based low-cost adsorbents for the removal of heavy metals (e.g., Cd, Cr, Cu, Ni, and Zn) from wastewater. The low-cost adsorbents derived from agricultural waste showed excellent performances in removing Cd, Cr, Cu, Ni, and Zn from wastewater. Using simulated wastewater, Babel and Kurniawan (2004) found that coconut shell charcoal (CSC) treated with nitric acid had a Cr adsorption capacity of 10.88 mg/g with a Cr concentration of 20 mg/L, the cost of CSC being approximately US$0.25/kg. At an initial Cr concentration of 1000 mg/L, the hazelnut shell activated carbon removed 170 mg/g (Kobya, 2004); however, at an initial concentration of 301.6 mg/L, ethylenediamine-modified rice husk removed 23.4 mg/g of Cr (Tang et al., 2003). At an initial concentration of 1000 mg/L of Ni, orange peel-based adsorbents removed 158 mg/g (Ajmal et al., 2000); at an initial concentration of 25 mg/L, the Ni removal was 6.01 mg/g (Annadurai et al., 2002). The adsorption capacities for Cu, Cd, and Zn were also higher at higher initial concentrations (Kurniawan et al., 2006). Kumar et al. (2011) demonstrated the efficiency of immobilized *Trichoderma viride* biomass as a biosorbent for the removal of Cr, Ni, and Zn ions from aqueous solutions and electroplating wastewater in a fixed-bed column. The initial concentrations of Cr, Ni, and Zn were 34 and 47, 28 and 190, and 24 and 239 mg/L, respectively. It is to be noted that the initial concentrations of these metals in drinking water are likely to be much lower than the typical wastewater concentrations; thus, these methods may need to be improved upon to achieve high binding affinity.

To remove As from drinking water, many metal-based adsorbents, including iron oxide-coated sand, ferrihydrite, red mud, activated alumina, TiO_2, $FePO_4$ (amorphous), MnO_2, MnO_2-loaded resin, gibbsite, goethite, kaolinite, zirconium-loaded activated carbon, natural zeolites (e.g., clinoptilolite), clinoptilolite–iron oxide, Portland cement, iron oxide-coated cement, titanium dioxide-loaded Amberlite XAD-7 resin, and iron (III)-loaded chelating resin have been used (Parimal, 2015). Activated carbon and activated alumina are the two most widely used adsorbents for removing As in drinking water. The synthesis of alumina following the conventional processes is relatively costly (Parimal, 2015). Recently, the synthesis of low-cost alumina was reported following the partial thermal dehydration method (Sen and Pal, 2009). In this method, the gibbsite precursor was processed to achieve an area of 335–340 m^2/g of alumina-based adsorbents. A column was filled with the activated alumina, which removed As from water to a level below 10 μg/L for more than 6000 bed volume water (Parimal, 2015). The annualized operating cost for PoU application of activated alumina was estimated to be US$1.0 per m^3 of water (Parimal, 2015).

In the presence of chlorine, chemical precipitation using alum, iron, and lime softening processes removed approximately 90% of As from water at initial As concentrations of ≥300 μg/L (Parimal, 2015). Arsenate [AsO_4^{3-}] and arsenite [AsO_3^{3-}] are the two commonly available forms of As in drinking water, which are mainly converted into arsenic sulfide, ferric arsenate, or calcium arsenate in these processes. These are then precipitated, with pH playing a crucial role. The fine colloidal As compounds (1–100 μm), formed by these processes, are separated through coagulation and settling followed by rapid sand filtration (Parimal, 2015). Simultaneous removal of several metals is possible in these processes. The cost of treatment has been estimated to be approximately US$ 0.89 per m^3 of water. Although these processes are effective for large-scale treatment of water with high concentration of As, these are not suitable for achieving the target level of 10 μg/L of As in drinking water as recommended by the regulatory agencies (e.g., WHO, 2011; USEPA, 2015). However, these processes may be used in conjunction with other methods (e.g., followed by low-cost adsorption) to satisfy the regulatory target.

In Bangladesh, a lower middle-income country (World Bank, 2016), rural populations apply a number of low-cost techniques to remove As from drinking water. Ahmed (2004) summarized few low-cost techniques used in South and East Asian countries (Table 4). The costs for the in-house and community-level techniques varied in the ranges of

Table 4
Low-cost techniques for As removal used in South and East Asian Countries.

Method suitable for	Process	Life (Year)	Cost (US$/m^3)	Annual capacity (m^3)
In-house application	Coagulation–filtration	3	1.7	16.4
	Iron-coated sand/brick dust	6	0.73	16.4
	Iron filling	5	0.24	16.4
	Synthetic media	5	1.84	16.4
	Activated alumina	4	2.39	16.4
Community application	Coagulation–filtration	10	1.21	246
	Granulated ferric hydroxide/oxide	10-15	1.2	820–900
	As–Fe removal (air oxidation–filtration)	20	0.054	730,000
	Activated alumina	10–15	3.2	164–200
	Reverse osmosis	10	3.72	328
	Ion exchange	10	3.4	25

US$ 0.24–2.39 and US$ 0.054–3.72, respectively, for 1.0 m^3 of water (Table 4). However, with the exception of activated alumina, ion exchange, and reverse osmosis, it is often difficult to remove As to a level below 10 μg/L (WHO, 2011; Parimal, 2015). The cost for these methods is high (Table 4). The cost of the activated alumina-based method used in rural Bangladesh was higher than that of Parimal (2015). By following the partial thermal dehydration method, the cost of alumina-based activated carbon can be significantly reduced (Parimal, 2015), which may be able to remove As from drinking water to a level below 10 μg/L (Parimal, 2015). Ferric chloride coagulation followed by filtration through iron-coated sand has been found to efficiently remove As from drinking water. Ali et al. (2001) applied this technique, which was found to remove As to a level below 20 μg/L from tube well water with a maximum As concentration of 400 μg/L. The technique is simple and can be built in house, and the overall cost is estimated to range between US$ 0.73 and 1.21 per m^3 of water. In addition, few methods are also in practice, which often face difficulties in reducing As to a level below 10 μg/L. Some of these methods employ bucket treatment units, Stevens Institute Technology, fill-and-draw treatment units, tube well-attached arsenic treatment units, and iron–arsenic treatment units. These methods follow the principle of coagulation, sedimentation, filtration, and adsorption (Ahmed, 2004). Through future research, the performance of these methods may be improved at a relatively lower cost.

To compare the different methods, Table 5 presents the findings from few major treatment methods for heavy metal removal. At least 80% of Cd was removed through coagulation, ion exchange, precipitation softening, and reverse osmosis (RO) membranes (Table 5). More than 80% of Hg was removed by coagulation, precipitation softening, RO membranes, and use of granular activated carbon (GAC) (WHO, 2011). The ion exchange and activated alumina adsorption treatments were effective in removing U, whereas ≥80% of As and Se were removed by ion exchange, activated alumina, and RO membranes (Table 5). Pb was effectively removed by coagulation, low-cost and commercial activated carbon, and alginate beads. However, the ion exchange, membrane filtration, electrodialysis, membrane distillation, and hybrid methods are likely to be more expensive than physical–chemical removal and low-cost adsorbents (Pianta et al., 2000; Parimal, 2015).

The presence of As in groundwater has become a pressing issue worldwide (Camacho et al., 2011; Hashim et al., 2011; ATSDR, 2015). In 2003, >50 million people in Bangladesh consumed drinking water with As levels exceeding 50 μg/L (Mondal et al., 2006). In the Southern Vietnam and Cambodian borders, about two million people were at a risk of chronic As poisoning (Buschmann et al., 2008). In Mexico and Latin American countries, at least four million people consumed drinking water with toxic levels of As (Bundschuh et al., 2006). At least 30 countries have reported chronic As toxicity from consumption of drinking water (Chowdhury et al., 2000), most being low- or medium-income countries (Fig. 1). Approximately 671 and 2950 million people in 31 and 51 countries belong to the categories of low-income and lower middle-income groups, respectively (Fig. 1). A total of 2239 million people from 53 countries belong to the upper middle-income group (World Bank, 2016). More than 100 million people belonging to these groups (e.g., India, Bangladesh, Pakistan, Nepal, Cambodia, Vietnam, Lao PDR, Mali, Nigeria, Nicaragua, Mexico, Peru, etc.) are facing problems due to high As levels in groundwater. Many of these populations cannot afford commercially available bottled water or advanced treatments at small/rural community or individual levels. For instance, Bangladesh is a low–medium-income country with 31.5% of the population living below the poverty line (World Bank, 2016; CIA, 2016), many living in rural areas. These people often do not have the access to As-free drinking water (Chakraborti et al., 2010). A similar situation prevails in some parts of many countries (e.g., Mexico, India, Vietnam, China, and El Salvador). In 2012, 12.7% of the world's population (896 million) lived on US$1.90 a day or below; conversely, in developing countries, >2.1 billion people lived on less than US$3.10 a day (CIA, 2016). These populations are unlikely to be able to afford these expensive advanced technologies (World Bank, 2016).

There is a need to develop low-cost technologies for individual-level and small community-level applications. PoU treatments may be considered for small community- and individual-level applications. The PoU treatments include an adsorption filter or an ion-exchange system, which can be installed to a single or multiple taps. Some PoU treatments can have the RO unit attached to a potable water storage tank connected to a dispensing faucet (Parimal, 2015). The columns, filled with partial thermal dehydrated alumina-based activated carbon, have shown excellent performance in removing As from drinking water at a relatively low cost, which can be studied for use in PoU filters with further improvement (Thomson et al., 2003; Kommineni et al., 2003). The locally applied low-cost methods in the South and East Asian countries (Table 4) must be improved upon to bring As to a level below 10 μg/L at a relatively low cost. In addition to As, the PoU treatments can remove few other heavy metals. Some PoU treatments have been successfully used: for instance, activated carbon for Pb removal (Sublet et al., 2003); carbonate minerals for Cu and Pb removal (Andy, 1996); zeolites for Pb removal (Sublet et al., 2003); and the binary mixture of acid- and steam-based GAC for Cu, Pb, and Zn removal (Ahmedna et al., 2004). Further research is needed to improve these PoU treatments and lower the cost for individual use.

Table 5
Methods for removing heavy metals from drinking water.

r	Coagulation	Ion exchange	Precipitation softening	Activated alumina	Reverse Osmosis Membranes	Activated carbon	Low-cost activated carbon	Alginate beads
Arsenic	++	+++	++	+++	+++			
Cadmium	+++	+++	+++		+++			
Mercury	+++		+++		+++	+++		
Selenium	++	+++		+++	+++			
Uranium	++	+++	++	+++				
Lead	+++		++			+++	+++	+++

++: ≥50% removal; +++: ≥80% removal.

In recent years, carbon nanotube (CNT)-based adsorbents have shown excellent performance in removing several heavy metals including Cr (Di et al., 2006), Pb (Li et al., 2005), Zn (Purnachadra et al., 2007), and As (Peng et al., 2005) from drinking water. However, the large-scale use of CNT might be limited by their high cost and toxicity (Upadhyayula et al., 2009). The cost of CNT adsorbents was reported to be in the range of US$25–38 per kilogram. Dalton et al. (2003) reported that the cost of CNT could be reduced to $10 per kilogram if commercial quantities were produced by a specialized corporation using appropriate technologies and optimized parameters. It is imperative that the cost and toxicity of CNT be reduced prior to large-scale use in PoU treatments.

5. Future research needs

In the past, the effects of consuming heavy metal-contaminated drinking water were characterized and technologies were developed to remove metals from drinking water. To ensure that public health is better protected, future study is needed to address the following issues:

- Co-exposure to multiple heavy metals can have synergistic or antagonistic effects on human health, and the NOAEL can be greatly affected during co-exposure to multiple metals. In particular, As in groundwater has become a global issue. In many cases, the co-presence of As, Mn, and/or Sb in drinking water has been reported. Co-exposure of As with other heavy metals (e.g., Sb and Mn) must be better characterized in the context of the effects on human health. Regulatory agencies may link the maximum limits of a metal in drinking water to the co-exposure scenarios as well as other factors (e.g., epidemiological and toxicological effects, treatability, etc.).
- Leaching of heavy metals from WDSs and plumbing system can be significant. The release of metals is likely to be influenced by several factors, including pH of water, temperature, residence time, pipe materials, disinfectant type and dose, and age of WDS pipes. Future study may better explain the effects of these factors and their interaction effects on heavy metal leaching.
- For small/rural communities and individuals, PoU treatments might be an alternative to ensure safe drinking water. Past studies have demonstrated that few low-cost adsorbents, when filled into a column, reduced As, Cu, and Pb significantly, whereas As was reduced to below the WHO guideline value of 10 μg/L at a bed volume of ≥6000. The cost was estimated to be low. In addition, few locally available low-cost technologies have been applied in the rural areas of South and East Asian countries. In future, research is needed to improve the low-cost adsorbents focusing on their in-house applications.
- Adsorption-, precipitation-, and filtration-based technologies are likely to generate significant amounts of heavy metal-contaminated wastes. Safe disposal of sludge, saturated media, and liquid wastes rich in heavy metals is a matter of concern. Several methods can be tested to address this issue. One such technique includes blending heavy metal-contaminated materials into stable waste or engineering materials, such as glass, bricks, concrete, or cement blocks, followed by safe disposal. Another option is to assess the reduction of heavy metals from aqueous solutions using the low-cost materials. Past studies have noted significant reduction of heavy metals through biochemical processes in the presence of cow dung (Ali et al., 2001). Recently, the sequestration of heavy metals has been demonstrated using natural zeolites (Diale et al., 2011). Future study is needed in these directions.
- CNT-based adsorbents have shown excellent performance in removing heavy metals (Cr, Pb, Zn, and As), bacteria, and viruses, which can be the excellent adsorbents in PoU treatments. The primary inhibiting factors are the cost and potential toxic effects. Research is needed to reduce the cost and toxic effects of CNT-based adsorbents, so that these materials can be used in the PoU treatment.
- Protonated alginate beads have shown promising results in removing Pb from water at relatively higher initial concentrations. Their performance at low initial concentrations of Pb and other heavy metals needs to be investigated. Further, efforts to minimize the cost of water production are needed.

6. Conclusions

This paper investigated the state of research on heavy metals in drinking water to date. The paper focused mainly on the occurrences and variability of heavy metals in drinking water, their sources, exposure, health effects, treatment, and the critical gaps that are need to be filled to supply safe drinking water. The literature to date has demonstrated that exposure to heavy metals through consumption of drinking water is almost unavoidable. In particular, people in small and rural communities and individuals are vulnerable. Exposure to few heavy metals (e.g., As, Cd, and Pb) is a pressing issue due to the significant risks to human health. Co-exposure to multiple heavy metals must be carefully studied for synergistic or antagonistic effects.

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