



Arsenic contamination in groundwater: An overview of emission sources, concentration control, and removal methods

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ABSTRACT

Arsenic is a chemical element with the symbol As and atomic number 33 in the fifteenth and fourth periods of the periodic table. It is the twentieth most abundant element in the earth's crust, the twelfth in the human body, and the fourteenth in seawater. Arsenic contaminants in groundwater in the world include four aquifers in delta basins and alluvial plains, inland basins in arid and semi-arid regions, mining in areas with sulfide minerals, and geothermal sources. Researchers estimate that more than 500 million people worldwide are exposed to arsenic, and the worst type of groundwater contamination ever reported was in Bangladesh, where about 80 percent of the population is affected. Sources of arsenic contamination include natural sources such as rocks weathering, fossil fuels, etc., and human resources include drainage of acid mines, paint factories, glass production, etc. Controlling the concentration of this element in groundwater can be influenced by various factors such as oxidation and reduction (Eh), solution composition, type of sediment mineral, microbiological activity, etc. Methods of removing this contaminant include biological methods using bacteria and adsorbents such as titanium dioxide.

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1. Introduction

Arsenic is the chemical element marked with the symbol As in a periodic table, also with atomic number 33 in the fifteenth and fourth periods of the periodic table as a metalloid is directly below phosphorus and is similar to phosphorus in terms of physicochemical properties. Arsenic is the twentieth most abundant element in the earth's crust, the fourteenth most abundant element in seawater, and the twelfth most abundant element in the human body (Pal, 2015). The concentration of arsenic in the earth's crust is between 0.5 to 2.5 mg/kg and it is possible that its concentration in clay-like sediments can reach more than 15 mg/kg. Also, the concentration of arsenic in igneous rocks is between 1.5 to 3 mg/kg and in sedimentary rocks, it ranges between 1.7 to 400 mg/kg (average 3 mg/ μ g). Arsenic is produced in large quantities in countries such as the United States, China, and Chile.

Arsenic can be captured from gold, lead, and copper smelting dust, also from roasting arsenopyrite grains with a chemical formula FeAsS, the most abundant arsenic ore (Kabata-Pendias and Szteke, 2015). Organic and inorganic compounds of arsenic forms may be present in the environment in various states of oxidation (-3, 0, +3, and +5). In natural waters, including groundwater, arsenic is mostly present in the mineral forms arsenite (trivalent) (H_3AsO_3) and arsenate (Five-valent) (H_3AsO_4), both of which are classified by the World Health Organization (WHO) as compounds. They are known to be toxic and carcinogenic. Mineral compounds of arsenic include hydrogenated compounds (such as arsenic), halides (such as fluorine), oxides, acids, and sulfides (Pal, 2015). The organic compounds of arsenic are mostly stored in the body of marine animals and it is called arsenobetaine with the chemical formula $C_5H_{11}AsO_2$ (Ohta et al., 2004).

Arsenic may be present naturally in 200 minerals, including arsenite, arsenate, arsenide, elemental arsenic, sulfides, and oxides of varying degrees, with the highest concentrations of metal oxides and sulfide minerals, In particular iron oxides. The difficulty of arsenic contamination in groundwater can be the consequence of sites with high concentrations of these minerals if

geochemical conditions of arsenic release from these minerals are provided. Conditions of geochemical including pH, aerobic or regenerative environment, groundwater flow, and transfer are widely used as causes of groundwater contamination with arsenic (Pal, 2015). The concentration of arsenic in different parts of the environment is shown in Table 1 (Ahuja, 2008).

Table 1. Arsenic concentrations in different parts of the environment

Environment	The range of Arsenic concentration
Air (ng/m ³)	1.5-53
Rainfall from ground air (µg/l)	0.46
Precipitation from unpolluted ocean air (µg/l)	0.019
River (µg/l)	0.2-0.264
Groundwater (well) (µg/l)	1-1000
Lake (µg/l)	0.38-1000
Sea water (µg/l)	0.15-6
Soil (mg/kg)	0.1-1000
Lake sediments (mg/kg)	2-300
River sediments (mg/kg)	5-4000
Sedimentary rock (mg/kg)	0.1-490
Igneous rock (mg/kg)	0.3-113
Metamorphic rock (mg/kg)	0-143

Groundwater arsenic contamination is a global problem and millions of people around the world are at risk of arsenic contamination and its compounds, which are known to be toxic and carcinogenic compounds (Singh et al., 2007). Numerous researches have been done in this field, which can be mentioned. Berg et al. (2001) in Vietnam studied arsenic contamination in groundwater and drinking water and its effect on human health and considered this contamination a threat to human health and the occurrence of chronic diseases. Chakraborti et al. (2016) conducted a study of the risk of arsenic contamination in groundwater and its effect on human health in several regions of India. In this study, an effect such as arsenic skin lesions, effects on hair, nails, and urine, as well as severe skin effects was identified in pregnant women on the health of individuals in the study areas. Ghosh et al. (2020) examined spatial modeling and mapping of groundwater arsenic contamination and risk assessment through spatial interpolation techniques in West Bengal. The results of this study showed that the ineffective blocks in the pre-monsoon season (March-April-May) of 2006 were significantly affected at the end of 2008. The regression model predicts that if this trend continues, after ten years, 2/3 blocks of these districts will be affected by arsenic. Cuong et al. (2022) investigated an integrated active biochar filter and a capacitive ionization system to remove

high-performance arsenic from groundwater. In this study, manganese dioxide biochar composite filter-rice husk was used for oxidation of As (III) to As (V) and initial removal of As (III, V). They concluded that an integrated system could remove arsenic from real groundwater to achieve the value of WHO guidelines for drinking water quality. Therefore, the purpose of this article is to investigate the sources of arsenic contamination in groundwater and to provide methods to remove this pollutant.

2. Results and discussion

2.1. Arsenic contamination aquifers in groundwater

Arsenic contamination in groundwater has been reported in several countries, in terms of the geographical distribution of arsenic-containing aquifers is divided into four categories, which are:

1. Delta basins and alluvial plains, which include India, Bangladesh, Nepal, Taiwan, Colombia, Laos, Vietnam, China, Hungary, and Romania.
2. Inland basins of arid and semi-arid regions, which include Mexico, Chile, Argentina, Spain, and the United States.
3. Mining in areas with sulfide minerals, including the United States, China, Ghana, Mexico, Brazil, Australia, the United Kingdom, and Portugal.

4. 4. Geothermal resources that include the United States, Japan, New Zealand, Chile, Argentina, Ecuador, France, and Italy (Alloway et al., 2013).

2.2. Arsenic contamination in groundwater in different countries of the world

Natural arsenic in groundwater affects millions of people around the world. Arsenic contamination in groundwater has been found in various countries of the world including Bangladesh, Argentina, Chile, Mongolia, Canada, Nepal, Taiwan, USA America, Poland, Pakistan, Vietnam, China, India, Thailand, Ghana, Laos, Mexico, Hungary, UK, and Cambodia. Even in developed countries such as the United States, nearly 10 percent of the country's groundwater resources contain more than standard arsenic in groundwater. According to the WHO, the standard arsenic in groundwater was 10 µg/l in 1993. This standard was last updated in 2006 by the United States. However, a concentration of 50 µg/l is the maximum permissible concentration of acceptable contamination in Bangladesh. More than 100 million people in Bangladesh are influenced to arsenic contamination in groundwater, according to the WHO. Some experts estimate that more than 500 million people worldwide are exposed to the infection. The worst type of arsenic contamination in groundwater was ever reported in Bangladesh in the 1970s at a depth of 10 to 40 meters. In general, the level of arsenic contamination in groundwater in Bangladesh is very high and approximately 80% of the population is exposed to this contamination. Undoubtedly, the source of this contamination in Bangladesh is of natural and geogenic, which includes the trivalent form of arsenic, between 50 to 60% of the total arsenic contamination in different parts of the country (Ahuja, 2008; Podgorski and Berg, 2020).

2.3. Sources of Arsenic Contamination in Groundwater

Arsenic Contamination in Groundwater in countries that use arsenic-contaminated groundwater as a major source of drinking water, there are serious risks to human health. Arsenic emissions from various natural and human sources can contaminate groundwater. Natural resources include weathering of rocks,

fossil fuels, and marine sedimentary rocks, various minerals, and volcanic activity in igneous rocks. Among the human resources of arsenic that cause groundwater contamination can be acid drainage, coal combustion, paint factory, glass factory, use of arsenic as a wood preservative, and use of insecticides. The use of arsenic-contaminated groundwater for irrigation of agricultural lands also causes the accumulation of arsenic and its toxic elements in the soil. Arsenic accumulated in the soil is then absorbed by plants and consumed by animals, and thus enters the food chain (Ahuja, 2008; Santha et al., 2022).

2.4. Control of arsenic concentration in groundwater

A variety of geochemical and geological conditions can control the concentration of arsenic in groundwater. These factors include leachate of geological materials, pH, oxidation and reduction (Eh), solution composition, type of sediment mineral, the particle size distribution of groundwater aquifers, type of groundwater aquifer mineral, water hydraulic system underground, the extent of microbiological activity and reactions related to the biogeochemical activity (Ahuja, 2008; Becher Quinodoz et al., 2019).

2.5. Arsenic under oxidation and reduction conditions

The mobility of arsenic in solution is significant especially under oxidizing conditions at high pH and under severely reducing conditions (Alloway et al., 2013). Arsenic can also be motile in freshwater samples in pH range from 6.5 to 8.5. Arsenic is also sensitive to changes in oxidation and reduction in groundwater. Unlike many oxy anions, arsenic is more mobile in reducing conditions (Datta, 2015). Arsenite is considered more toxic in the oxidation state. However, most recent studies have shown that arsenate can be reduced to arsenite. Therefore, exposure to both forms of arsenite and arsenate can be considered toxic compounds. Arsenate is stable in the oxidizing environment. For normal pH values in groundwater, arsenate species are predominant in solution with pH values of 2.2 to 6.9 as well as values between 6.9 and 11.5. Arsenite is moderately stable in the regeneration environment. The oxidation

rate of arsenite is greatly increased by the presence of other oxidants such as manganese oxides. Arsenite is typically more mobile in the acidic environment than arsenate and less

mobile in the alkaline environment (Welch and Stollenwerk, 2003). Figure 1 shows the different forms of arsenic at different pHs and oxidation and reduction conditions.

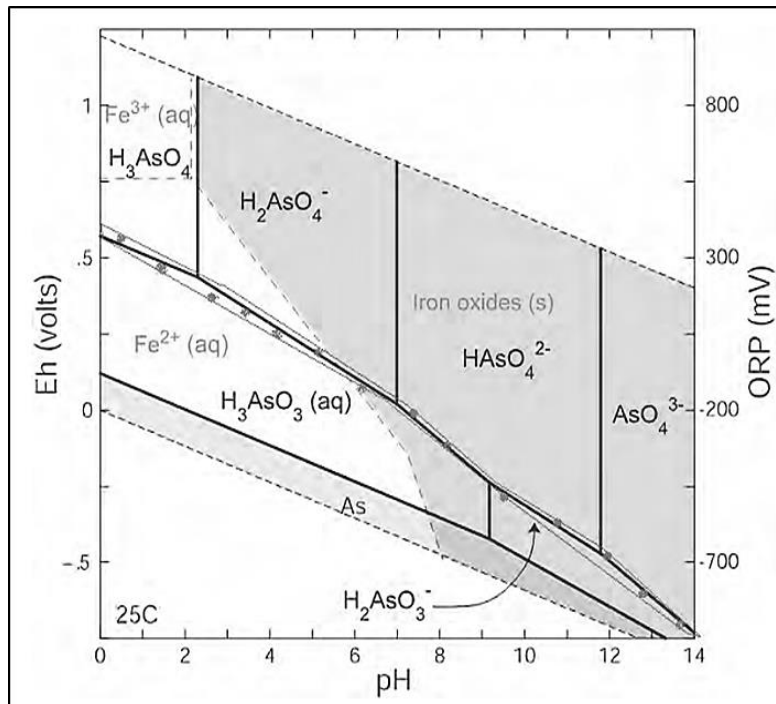


Fig. 1. Different forms of arsenic at different pHs and oxidation and reduction conditions

2.6. Modeling of arsenic transfer in groundwater

Prediction of the temporal and spatial distribution of arsenic in groundwater is obtained by pairing adsorption models with solute transfer models. Experimental adsorption models are used to model the transfer of reactive solutes in groundwater. The main benefit of these models is the simplicity of their mathematical calculations, which allow them to easily participate in solute transfer models. However, the ability of these models to describe arsenic adsorption in complex geochemical systems is limited. The chemical reactions for each cell in the model are solved separately using the geochemical model. Arsenic compounds are then transferred by the simultaneous transfer model. This procedure is repeated consecutively to the end of the simulation. Many models have been developed for the transfer of arsenic in groundwater. These models include Tranql, Hydrogeochem, Phast, Mst1d, Moc Phreeqc, and Phreeqc. These models have been used successfully for arsenic research in groundwater (Welch and Stollenwerk, 2003). In a study by Ghomi Avili and Makaremi (2020), the model of the spatial

and temporal distribution of pollutant concentrations in soil columns and groundwater contamination of the Gorgan plain of Iran was investigated using the LEACHM simulation model. The model used similar mathematical and numerical solution schemes to simulate the vertical motion of water and chemicals. Soil and groundwater of Golestan province are contaminated with arsenic as a toxic element, and spatial and temporal distribution of arsenic in the soil column have been estimated. Also, to develop a one-dimensional dynamic simulation model, it approximated the concentration of arsenic in the soil column and groundwater system by the finite differential (FDM) method in the unstable state of water flow where microbial activity and plant growth exist. The variations between the measured data and the simulation results were strongly influenced by the absorption coefficient. The simulation results showed that with increasing the total time from the beginning to the end of the simulation, the concentration of arsenic compounds approached the measured values.

2.7. Methods of arsenic removal in groundwater

Due to the fact that excessive concentrations of arsenic in groundwater are considered as a contamination and environmental problem for this water source, it should be eliminated using appropriate methods. Here are two ways to remove arsenic from groundwater.

2.7.1. Removal of arsenic from groundwater using biological methods

Among the biological methods is the use of bacteria to remove arsenic from contaminated groundwater. Which can be referred to a study conducted by Kao et al. (2013). In this study, the bacterial oxidation efficiency of arsenite (Trivalent arsenic) and then the removal of arsenate (Five-valent arsenic) from contaminated groundwater was investigated using bacterial biomass. The oxidizing bacterium arsenite (7325As) was taken from the groundwater aquifer of a native area of Taiwan that contained Blackfoot disease. Arsenite was then oxidized to arsenate and its removal process was investigated using arsenite oxidizing bacteria. Bacterial cells that were freeze-dried were responsible for absorbing arsenate. Dried bacterial cells (5 g/l) were added to 30 ml of the sample at concentrations of 500 and 1000 µg/l arsenate. The results showed that more than 99% of arsenate was removed from a sample containing arsenic-contaminated groundwater within 6 days, and concluded that the use of bacteria as arsenate adsorbents was a suitable biotechnology for the removal of arsenic from Groundwater is polluted.

2.7.2. Removal of arsenic from groundwater using titanium dioxide sorbent

In a study by Bang et al. (2005), the removal of arsenic from groundwater in New Jersey was performed using a granular (granular) titanium dioxide sorbent. In this paper, the kinetics and adsorption capacity of granular titanium dioxide for arsenate (Five-valent arsenic) and arsenite (Trivalent arsenic) were investigated. The adsorption capacities for the two forms of arsenic, arsenate and arsenite were 41.4 mg/g and 32.4 mg/g titanium dioxide, respectively. According to the system they designed, the suitable substrate for removing 39 micrograms per liter of arsenic was 3 liters of granular titanium dioxide. After passing the arsenic through this bed, the concentration of arsenic increased from 39 micrograms per liter to 10

micrograms per liter. They concluded that granular titanium dioxide adsorbent is very effective in removing arsenic from groundwater.

3. Conclusion

According to the contents of this study, the following results can be inferred:

- 1) Arsenic is a chemical element with the symbol As and is the twentieth most abundant in the earth's crust, the twelfth in the human body, and the fourteenth in seawater.
- 2) Arsenic pollution aquifers in different parts of the world include four aquifers in delta basins and alluvial plains, inland basins of arid and semi-arid regions, mining in areas with sulfide minerals and It is a source of geothermal energy.
- 3) The worst type of arsenic contamination is in Bangladesh, where approximately 80% of the population is exposed, which is the primary source of geogenic contamination. More than 500 million people worldwide are exposed to arsenic contamination.
- 4) Arsenic release in groundwater can be achieved through natural resources such as rock weathering, fossil fuels, marine sedimentary rocks, etc, and human resources such as acid mine drainage, coal combustion, dyeing plant, plant Production of glass, use of arsenic as a preservative for wood, etc.
- 5) The use of arsenic-contaminated groundwater for irrigation of agricultural lands has caused the accumulation of arsenic in the soil, which is absorbed by plants and after consumption by animals can enter the human food chain.
- 6) Various conditions including leachate, pH, oxidation, and reduction (Eh), solution composition, type of sediment mineralization, the particle size distribution of groundwater aquifers, type of groundwater aquifer mineral, groundwater hydraulic system, and microbiological activity, can control the concentration of arsenic in groundwater.
- 7) Models for predicting the temporal and spatial distribution of arsenic in groundwater are widely used include Tranql, LEACHM, Hydrogeochem, Phast, Mst1d, Moc Phreeqc, and Phreeqc.
- 8) Methods for the removal of arsenic contaminants in groundwater include the use of biological methods using oxidizing bacterial

biomass and the use of adsorbents such as TiO_2 .

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