



## Review

## Human health implications, risk assessment and remediation of As-contaminated water: A critical review



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## HIGHLIGHTS

- Arsenic is recognized as a Class-A human carcinogen.
- Groundwater As contamination has affected over 200 million people worldwide.
- This paper reviews current knowledge regarding As in the environment.
- A critical assessment of remediation of contaminated water is presented.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Arsenic (As) is a naturally occurring metalloid and Class-A human carcinogen. Exposure to As via direct intake of As-contaminated water or ingestion of As-contaminated edible crops is considered a life threatening problem around the globe. Arsenic-laced drinking water has affected the lives of over 200 million people in 105 countries worldwide. Limited data are available on various health risk assessment models/frameworks used to predict carcinogenic and non-carcinogenic health effects caused by As-contaminated water. Therefore, this discussion highlights the need for future research focusing on human health risk assessment of individual As species (both organic and inorganic) present in As-contaminated water. Various conventional and latest technologies for remediation of As-contaminated water are also reviewed along with a discussion of the fate of As-loaded waste and sludge.

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## 1. Introduction

Arsenic is a naturally existing metalloid and a carcinogenic element. The background natural concentration of As in the soils is  $\sim 5\text{--}10\text{ mg kg}^{-1}$  (Smedley and Kinniburgh, 2002; Basu et al., 2014); however, its distribution is not homogenous in the Earth's crust and is concentrated in certain geologic settings (Ravenscroft et al., 2009; Rahman and Hasegawa, 2011; Shah, 2014). Arsenic concentration in groundwater ranges from 0.5 to 5000  $\mu\text{g L}^{-1}$  and contamination from natural sources has been reported for >105 countries (Ravenscroft et al., 2009; Kippler et al., 2016).

The major sources of As in groundwater are minerals such as realgar (AsS), arsenopyrite (FeAsS) and orpiment ( $\text{As}_2\text{O}_3$ ). Overall, As sources are attributed to both natural processes, such as oxidative/reductive dissolution of As containing compounds sorbed onto pyrite minerals and anthropogenic activities such as use of pesticides, irrigation with As-contaminated water, semi-conductor manufacturer, phosphate fertilizers, mining and smelting activities, burning of coal, and timber preservatives (Shakoor et al., 2015; Shakoor et al., 2016).

Arsenic occurs in four oxidation states, i.e. arsenite (As(III)), arsenate (As(V)), elemental As ( $\text{As}^0$ ), and arsine (As(III)) (Sharma et al., 2014a). The solubility and mobility of As largely depends on pH and redox conditions, and is present in both organic and inorganic forms in the groundwater. Arsenite predominates in a reduced environment (deep groundwater) at a high pH value, while As(V) species prevail in an oxidized environment (shallow groundwater) at low pH (Abdallah and Gagnon, 2009; Bundschuh et al., 2012; Jackson et al., 2012).

In aqueous environments, As(V) exists as weak triprotic acid and it has different acid dissociation constants (pK) such as 2.20 and 6.97–11.53. At natural pH range (4–8) such as in groundwater, As(V) species are present as  $\text{H}_2\text{AsO}_4^-$  and  $\text{HAsO}_4^{2-}$ . Arsenate species prevail as an

amphiprotic material at pH 2.24–11.50, and thus it accepts or donates protons ( $\text{H}^+$ ) depending upon the variation in the pH of the aqueous environment (Mandal and Suzuki, 2002; Naidu et al., 2006a; Markley and Herbert, 2009).

On the other hand, As(III) occurs as a hydroxo-acid and has three dissociation constants (9.22, 12.13 and 13.40). Since the protons ( $\text{H}^+$ ) present on the hydroxyl groups (OH) of  $\text{As}(\text{OH})_3$  do not possess a doubly bonded oxygen neighbor, As(III) is considered to be a very weak acid and exists as a neutral or non-ionic species ( $\text{H}_3\text{AsO}_3$ ) in most of the groundwater conditions. Arsenite also predominates as both the amphiprotic and the polyprotic acid at pH > 9.22 in aqueous environment (Naidu et al., 2006a).

Organic As is also present in the environment as monomethylarsonic acid  $\{\text{CH}_3\text{AsO}(\text{OH})_2$ ; MMA), dimethylarsinic acid  $\{(\text{CH}_3)_2\text{AsOOH}$ ; DMA), trimethylarsine oxide  $\{(\text{CH}_3)_3\text{AsO}$ ; TMAO), arsenosugars (AsS), arsenobetaine (AsB), arsenocholine (AsC) and arsenolipids etc. (Singh et al., 2015). Methylated As compounds occur in a small amount (Gan et al., 2014); however, they may also be present as a major component in the soil. Both the MMA and DMA (cacodylic acid) were used extensively as herbicides and pesticides in the past – DMA was also applied as a cotton defoliant (Diwakar et al., 2015).

Arsenic contamination of groundwater is a global public health and environmental risk for over 200 million people in the world (Rasool et al., 2015; Mishra et al., 2016). Groundwater in most of the countries also acts as a pathway for As to enter into the food chain via drinking, cooking, bathing, and irrigation of food crops with As-contaminated water (Abid et al., 2016). Thus, intake of As-contaminated water and food is a major pathway for As exposure to humans. Long-term exposure to As is reported to cause skin lesions, neurotoxicity, diabetes, cardiovascular diseases, and various kinds of cancers such as skin, liver, bladder, and kidney (Hsu et al., 2016; Rasheed et al., 2017).

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