



## Review

## Arsenic and antimony in water and wastewater: Overview of removal techniques with special reference to latest advances in adsorption



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## ABSTRACT

Arsenic and antimony are metalloids, naturally present in the environment but also introduced by human activities. Both elements are toxic and carcinogenic, and their removal from water is of unquestionable importance. The present article begins with an overview of As and Sb chemistry, distribution and toxicity, which are relevant aspects to understand and develop remediation techniques. A brief review of the recent results in analytical methods for speciation and quantification was also provided. The most common As and Sb removal techniques (coagulation/flocculation, oxidation, membrane processes, electrochemical methods and phyto and bioremediation) are presented with discussion of their advantages, drawbacks and the main recent achievements. Literature review on adsorption and biosorption were focused in detail. Considering especially the case of developing countries or rural communities, but also the finite energy resources that over the world are still dependent, recent research have focused especially readily available low-cost adsorbents, as minerals, wastes and biosorbents. Many of these alternative sorbents have been presenting promising results and can be even superior when compared to the commercial ones. Sorption capacities were accurately compiled for As(III,V) and Sb(III,V) species in order to provide to the reader an easy but detailed comparison. Some aspects related to experimental conditions, comparison criteria, lack of research studies, economic aspects and adsorption mechanisms were critically discussed.

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## 1. Introduction

In the second half of 20th century, great concern was observed with heavy metals pollution, since many industrial and environmental accidents occurred, alerting the world for the environmental questions. In the last decades, Arsenic (As) and Antimony (Sb) have received attention from researchers and authorities, and have been described as critical issues in many research articles.

Arsenic and antimony are both metalloids, having properties of metals and nonmetals, and belong to Group VA of the periodic table (third and fourth elements, respectively). These trace elements with similar chemistry and toxicity are naturally occurring and commonly present together (An and Kim, 2009; Lehr et al., 2007). Anthropogenic sources are also responsible for their presence in the environment. The significance to human health and the impact in environment have explained the recent increasing interest for

arsenic and antimony abundance, behavior and remediation. Arsenic and antimony are both considered pollutants of priority interest by the European Union (Council of the European Communities, 1976) and by the Environmental Protection Agency of the United States (EPA, 1982).

The World Health Organization (WHO) established, in 1993, 10 µg/L as the guideline value for arsenic in drinking water. This value was defined considering the treatment performance, analytical achievability (WHO, 2011) and the risks related to human health. Several countries (USA and EU countries) adopted 10 µg/L as legal standard, but others have retained the previous value (50 µg/L). Considering the overall abundance of this element in the environment, to reach this limit in drinking-water can be critical in many parts of the world (Smedley and Kinniburgh, 2002). For antimony, UE and USA limits are 5 µg/L and 6 µg/L, respectively.

Present paper briefly reviews the sources, distribution in natural waters, chemistry, speciation and removal techniques of arsenic and antimony from water and wastewaters. Special emphasis is given to adsorption and to the latest findings in the field of alternative low-cost adsorbents.

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## 2. Sources and distribution in natural waters

### 2.1. Sources of arsenic and antimony

Arsenic occurs naturally, ranks 20th in natural abundance (component of more than 245 minerals) and is present in human body in a total content between 3 and 4 mg (NAS, 1977). Arsenic occurrence in the free state is unusual; it is largely found in combination with sulfur, oxygen and iron (Brewster, 1994). It is mobilized through a combination of natural processes, such as weathering reactions, biological activity and volcanic emissions as well as through a range of anthropogenic activities (Smedley and Kinniburgh, 2002). Most environmental arsenic problems were caused from mobilization under natural conditions. However, anthropogenic sources as mining activity, combustion of fossil fuels, use of arsenical compounds in agriculture (insecticides, pesticides and herbicides) and in livestock feed are additional pathways for environmental arsenic problems. Mine tailings can be heavily polluted with arsenic, presenting levels in the range of 0.9–62 g/kg (Kim et al., 2002), acting as main sources of arsenic for groundwater and surface water.

Antimony is much less predominant in nature than arsenic, but its significance for human health and for the environment is equally important. Antimony and its compounds have distinct properties that can be used for a variety of purposes. Diantimony trioxide ( $\text{Sb}_2\text{O}_3$ ) is used as a catalyst in the production of polyethylene terephthalate (PET) and as a flame retardant in the production of plastics, textiles and rubber (Reimann et al., 2010). About 60% of antimony is consumed in flame retardants and 20% used in alloys (Biswas et al., 2009). Antimony is used in brake linings, semiconductor components, battery grids, bearing and power transmission equipment, sheet and pipe and in pigments for paints. It is also applied as additive in glassware and ceramics, as an active ingredient in the treatment of Leishmaniasis disease (Amarasiriwardena and Wu, 2011) and, as elemental Sb, in ammunition (Guo et al., 2009). Antimony contamination is found in areas affected by mining activities, copper smelters or power plants. A significant input of Sb into the environment occurs at shooting ranges, since most bullets contain substantial amounts of Sb (Johnson et al., 2005). Due to antimony use in auto brake linings and disks, Sb release, as antimony trioxide (a potential carcinogen), occurs during braking (Ceriotti and Amarasiriwardena, 2009). China has the most rich Sb resources in the world and plays an important role in global anthropogenic emissions, leading to severe environmental contamination (He et al., 2012).

### 2.2. Distribution of arsenic and antimony in natural waters

#### 2.2.1. Groundwater

Depending on the local availability, drinking-water can be produced from surface water, groundwater or rain water. Humans can be exposed to arsenic by specific occupational exposure, airborne particulate matter, food (fish and shellfish) and water ingestion. The greatest risk of arsenic exposure is probably related to drinking-water (WHO, 2011). Literature has reported a wide range of typical arsenic concentrations in groundwater, from  $<0.5 \mu\text{g/L}$  to 5 mg/L (Smedley and Kinniburgh, 2002; WHO, 2004), with many aquifers around the world presenting levels above  $50 \mu\text{g/L}$  (Mandal and Suzuki, 2002; Smedley and Kinniburgh, 2002). The occurrence and origin of arsenic in groundwater depends on various factors such as adsorption-desorption, precipitation-dissolution, oxidation-reduction, ion-exchange, grain size of sediments, organic content, biological activity and aquifer characteristics. Under natural conditions, it is in groundwater that the greatest ranges and highest As concentrations are found, due to the strong influence of

water–rock interactions and to the tendency in aquifers for physical and geochemical conditions favorable for As mobilization and accumulation (Smedley and Kinniburgh, 2002). If the groundwater is affected by any of the anthropogenic pollution sources (mining, agriculture and industry), arsenic levels can reach dozens of mg/L, although, in this case, the problem tends to be restricted to a definite area. Most notable arsenic problems in the world occurred in Argentina, Bangladesh, Chile, China, Hungary, India (West Bengal), Mexico, Romania, Taiwan, Vietnam, many parts of the USA, Nepal, Myanmar and Cambodia (Bhattacharya et al., 2007; Gan et al., 2014; Mandal and Suzuki, 2002; Sharma and Sohn, 2009).

Regarding to antimony, speciation and distribution in freshwater have not been extensively studied. Total Sb dissolved concentrations in groundwater have been reported in the range  $0.010$ – $1.5 \mu\text{g/L}$  (Filella et al., 2002a), but anthropogenic sources can be responsible for much higher levels.

Arsenic and antimony levels in geothermal groundwater (Izmir, Turkey) were reported in the ranges  $0.7$ – $170 \mu\text{g/L}$  and  $0.06$ – $26 \mu\text{g/L}$ , respectively (Aksoy et al., 2009). Groundwater in the vicinity of abandoned antimony mines in Slovakia presented As and Sb levels up to  $285 \mu\text{g/L}$  and  $1000 \mu\text{g/L}$ , respectively (Hiller et al., 2012).

#### 2.2.2. Surface waters

Baseline concentrations of arsenic in rivers have been reported in the range  $0.1$ – $2.0 \mu\text{g/L}$ , depending on the composition of the surface recharge, contribution from base flow, bedrock lithology and river flow (Smedley and Kinniburgh, 2002). Wider ranges can however be found due to naturally-occurring As, geothermal influence and anthropogenic causes, as documented in literature (Baeyens et al., 2007; Barats et al., 2014; Nath et al., 2009; Ritchie et al., 2013; Sanchez-Rodas et al., 2005; Singh et al., 2010; Smedley and Kinniburgh, 2002). Gomati River and its tributaries (Ganga Plain, northern India) presented As concentrations in the range of  $1.29$ – $9.62 \mu\text{g/L}$  (Singh et al., 2010). Arsenic levels of  $0.97$ – $3.6 \mu\text{g/L}$  were found for Zenne River, a sewage contaminated tributary of the Scheldt estuary (Belgium) (Baeyens et al., 2007). Alpine/Mediterranean Var River watershed (France) presented an extremely variable As concentration, ranging from  $0.1$  to  $263 \mu\text{g/L}$  (Barats et al., 2014). A remarked increase in these levels could be attained in areas influenced by mining as showed in Ashanti River, Ghana (Nath et al., 2009), Tinto and Odiel Rivers, in Spain (Sanchez-Rodas et al., 2005), with As levels ranging from  $0.5$  to  $7900 \mu\text{g/L}$ , and in water streams in Stampede and Slate Creek watersheds, Kantishna Hills mining district (Alaska, USA), with As concentrations as high as  $720 \mu\text{g/L}$  (Ritchie et al., 2013).

Lakes are also a possible source of fresh water for human consumption purposes. Arsenic concentrations in lake waters are usually close to or lower than those found in river water (Smedley and Kinniburgh, 2002). Baseline concentrations are typically below  $1 \mu\text{g/L}$  or near  $1$ – $2 \mu\text{g/L}$  (Azcue et al., 1994, 1995; Azcue and Nriagu, 1995; Ikem and Egilla, 2008; Mannio et al., 1995; Reuther, 1992), but values in the range  $1$ – $157 \mu\text{g/L}$  have been reported in lakes of different countries, as for example Lake Mohawk (USA), two lakes of China and in south east part of Sindh, Pakistan (Arain et al., 2009; Barringer et al., 2011).

The antimony distribution in natural waters has received much less attention than arsenic. Reimann et al. (2010) presented a comprehensive review of published data on Sb distribution in different environmental samples. Sb levels in freshwaters have been reported in the range from ng/l to a few  $\mu\text{g/l}$  (Reimann et al., 2010; Wang et al., 2011), with the average Sb concentration in world rivers being  $1 \mu\text{g/l}$  (Wang et al., 2011). Higher concentrations related to localized anthropogenic sources can be found. In Stampede and Slate Creek watersheds, Kantishna Hills mining district (Alaska, USA) Sb concentrations of  $239 \mu\text{g/L}$  were found (Ritchie

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