Article

Antimony removal from water and wastewater by different technologies: A review

Abdolmajid Fadaei
Department of Environmental Health Engineering, School of Health, Shahrekord University of Medical Sciences, Shahrekord, Iran
E-mail: ali2fadae@yahoo.com

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Abstract
Antimony (Sb) and its compounds, as global priority contaminants, are found in soils and water due to natural and anthropogenic sources. Elimination of Sb from water and wastewater is necessary because of its potential harm to the ecosystem and human health. This study aims to present different technologies used for Sb removal from water and wastewater. Various treatment techniques used for Sb removal, including reverse osmosis (RO), phytoremediation, photooxidation, electrodeposition, precipitation/coagulation, membrane filtration, electrocoagulation, biosorption, and adsorption, have been considered widely by researchers and revealed acceptable findings. Adsorption studies indicated that the adsorption capacity for Sb (V) and Sb (III) is 1.65-287.88 mg/kg and 3.11-250 mg/kg, respectively. The percentage of Sb removal from water and wastewater using coagulation and flocculation ranges from 71.02 to 100%. In this article, several methods for elimination of Sb are explained to understand how this process occurs and what study gaps are required to be discussed to enhance performance. Adsorption technology is the most extensively used method for elimination of Sb. Biological methods, namely phytoremediation, are also a promising technique, and further research is required in this regard. The choice of an appropriate method for a given area depends on the economical, environmental, and social conditions.

Keywords adsorption; wastewater; ground water; water treatment; Antimony (Sb) removal.

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1 Introduction

Over recent years, the elevated levels of antimony (Sb) pollution in water have produced global anxiety. Anthropogenic activities, such as mining waste and industrial activities are fairly accountable for the increased levels of Sb. Sb exists in natural waters in several oxidation states, including Sb (-III), Sb (0), Sb (III) and Sb (V), among which Sb (III) and Sb (V) are the two most common types of Sb. Different chemical states of Sb determine its mobilization and toxicity. Sb (III) is more toxic than Sb (V) (Luo et al., 2015; Wang et al., 2020a). Inorganic compounds of Sb are more poisonous than its organic forms and the Sb (III) compounds are anticipated to be nearly 10 times more toxic than Sb (V) oxo-anionic species(Smichowski, 2008). Sb trioxide (Sb_2O_3) is possibly carcinogenic to humans (group 2B), while carcinogenic impacts of Sb trisulfide (Sb_2S_3) on humans have not been recognized (group 3) with regard to the grouping of the International Agency for Research on Cancer (IARC) (Herath et al., 2017; Ungureanu et al., 2015). Antimony and antimonide are components of crust, the Sb's concentration in the crust is nearly 0.3 μg/kg, and its concentration in the upper and lower crust is alike (Gómez et al., 2005). The World Health Organization (WHO) has regulated the maximum admissible concentrations of Sb in consumable water and soil to be 0.020 mg/L and 36 mg/kg, respectively (Guo et al., 2009). The major sources of Sb, including natural weathering of Sb ore, mining, smelting, burning of fossil fuels, and wide use of Sb constituents, have increased the concentration of Sb in the geochemical environment, leading to air, water and soil pollution (Hu et al., 2017). Based on present report, approximately 80% of Sb generation was concentrated in China, Russia, and Bolivia (Fig. 1) (Bolan et al., 2022).

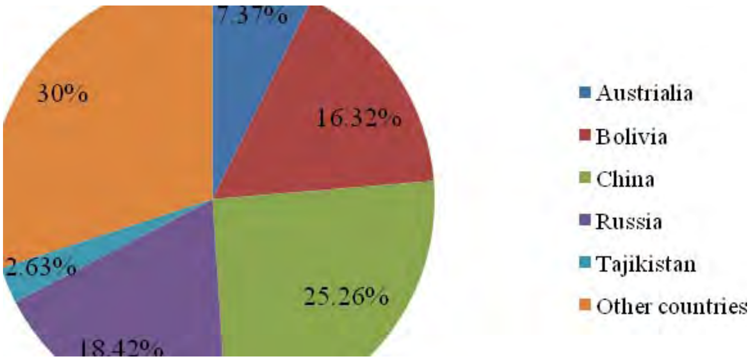

Fig. 1 Worldwide Sb production (Bolan et al., 2022)

China has the largest resources of Sb across the world. Great amounts of Sb are discharged from mining and smelting processes in the mine or near mine sites, causing severe Sb pollution (Hu et al., 2016). Besides, Sb can simply get into one's body through skin contact, respiration, and food chain. All Sb and Sb compounds have toxicity to the human body, but Sb (III) compounds are more mobile and bioavailable, and thus, more toxic to human and environment health than Sb (V) species (Li et al., 2018a). Negative effects of Sb on human health can be divided into the following two groups: acute effects and chronic effects. The acute effects of Sb include vomiting, muscle pain (myalgia), cramps, haematuria, pancreatitis, nephritis, abdominal colic, hepatic cirrhosis, diarrhea, and skin rashes. Among the chronic effects are cough, dyspnoea, pneumoconiosis, chronic lung changes, chronic bronchitis, early tuberculosis, and cardiotoxicity (Luo et al., 2015; Pierart et al., 2015). Many technologies like reverse osmosis, photooxidation, electrodeposition, coagulation/precipitation, and membrane filtration, electrocoagulation, biosorption, and adsorption have been used to eliminate Sb from water (Ahmadi et al., 2020; Fadaei, 2021; Taie et al., 2021; Wu et al., 2021a). Most of these methods are costly and/or inefficient in eliminating Sb from very dilute solutions. Adsorption is one of the best available treatment techniques for Sb elimination from water because it is an easy, safe, compact, simple to operate and greatly proficient method. The purpose of this study is to present different technologies used for removal of Sb from water and wastewater.

2 Antimony (Sb) Removal Mechanisms by Different Technologies

2.1 Adsorption

The adsorptive behavior of an adsorbent depends on the chemical form of the adsorbate. The Sb adsorbents are classified into inorganic adsorbents, organic adsorbents, compound adsorbents, etc. Various methods have been used for elimination of Sb, including activated carbons, Fe and Mn-based sorbents, Fe-Cu binary oxides, titanium oxides, biosorbents, magnetic adsorbents, activated alumina, and miscellaneous adsorbents.

2.2 Biological and bioremediation techniques

In biological systems, anaerobic and aerobic bacteria, algae, fungi, and plants are able to catalyze the removal of Sb under environmental conditions. Bioremediation is based on conversion of toxic, water soluble Sb oxyanions to elemental, water insoluble Sb. There are various biological treatments, including bioremediation, biosorption, biofiltration. Moreover, phytoremediation is a bioremediation process in which different types of plants are used to remove contaminants from the soil and groundwater. Different mechanisms of phytoremediation include phytofilteration, phytostabilization, phytovolatilization, phytoextraction.

2.3 Coagulation and flocculation

The coagulation-flocculation method is the process of adding mineral coagulants or natural polymers to water and wastewater to unstable contaminants. There are various coagulants, including ferric chloride, poly aluminum chloride, poly ferric sulfate, and ferrous sulfate.

2.4 Membrane filtration

In the membrane separation process, the use of semipermeable membranes, selectively permeable to water and certain solutes, allows for separating target particles from the solution. There are various membrane separation alternatives, including ultrafiltration, reverse osmosis (RO), nanofiltration and electrodialysis. The elimination efficiency of pollutant is affected by several factors in membrane treatment, such as the quality of membrane, component concentration, membrane electrical potential, flow rate, pressure gradient force, and chemical specifications of the water and wastewater (temperature, pH, dissolved organic matter, etc) (Long et al., 2020).

2.5 Photooxidation

Photocatalysis reduction is a relatively new technology for water and wastewater treatment and a suitable way to realize green chemistry. When the photocatalytic materials are subjected to the radiation energy equal to or greater than that of the photocatalyst's band gap, the electron-hole pairs are generated and they decompose into electrons (e^-) in the conduction band and holes (h^+) in the valence band. The e^- and h^+ cause reduction and oxidation of molecules adsorbed on the surface of photocatalytic pollutants (Rueda-Marquez et al., 2020).

2.6 Electrochemical treatment

This method is applicable for remediation of the industrial effluents and groundwater, removing different contaminants before releasing or recycling the treated water (Mousazadeh et al., 2021a). The elimination efficiency of Sb is affected by several factors in electrochemical treatment like the materials of electrodes, primary concentrations, pH, current density, duration, aeration rate, interfering ions, etc (Long et al., 2020).

The most important mechanisms of electrochemical treatment are as follows (Mousazadeh et al., 2021b):

Anode: $M \rightarrow M^{n+} + n\, e^-$ metal dissolution (1)

Cathode $2H_2O + 2\, e^- \rightarrow H_2 + 2\, OH^-$ hydrogen evolution and hydroxide formation (2)

$M^{n+} + n\, OH^- \rightarrow M(OH)_n$ metal ion hydrolysis (3)

3 Methods

This review has principally focused on methods and processes. Several papers on the topic were retrieved from databases, such as Google Scholar, Web of Science, and Science Direct. Keywords, such as "surface water", "biological technology," "**c**oagulation", "photooxidation", "flocculation", "electrodialysis", "bioremediation," "phytoremediation", "electrochemical method" "membrane technology", "wastewater," "ground water", "water treatment" "antimony removal", and "adsorption" were used to retrieve proper papers. After a thorough search and removing articles that were not directly related to Sb removal from water, a total of 131 original papers were identified as eligible to be included within the review. The review articles providing a perception of various mechanisms of each treatment method were excluded. Types of water and wastewater, such as surface water, geothermal water, groundwater, industrial wastewater, and mining wastewater were investigated in this research.

4 Results and Discussion

These articles used several techniques, including adsorption (16), bioremediation and biological method (9), coagulation and flocculation (7), membrane filtration (4), electrochemical method (3) photocatalytic process (2) and other technologies (4) (Tables 1-7). Different techniques have been used and proposed to eliminate Sb from aqueous phase. Considering legal limits and toxic effects, the removal of these metalloids from water and wastewater is mandatory due to anthropogenic or natural reasons.

4.1 Adsorption

During the course of the past few years, adsorption played a key role in elimination of heavy metals. Currently, a significant number of adsorbent materials have been developed for elimination of Sb from contaminated water. Adsorption is considered as one of the most attractive methods due to its simple use and easy operation, cost-effectiveness and low secondary contamination risk, and admirable handling in low concentration of wastewater (Long et al., 2020). According to this study, the most adsorption isotherms of Sb (III) and Sb (V) fit the Langmuir isotherm model (about 79%). Additionally, the maximum adsorption of 759 mg/g Sb (III) was sorbed by Fe-doped birnes sites, and 287.88 mg/g Sb (V) was sorbed by Zirconium-Based Metal-Organic Frameworks (Fig. 2 and 3).

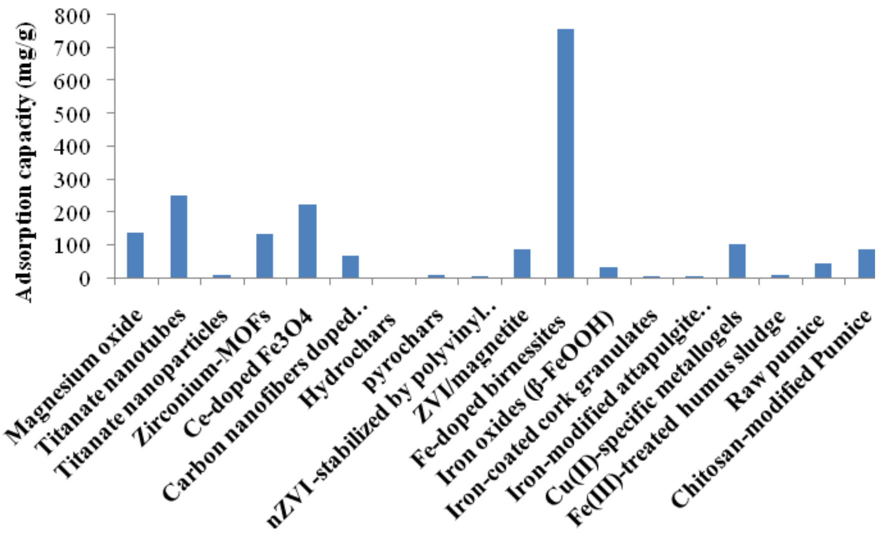

Fig. 2 Removal performance of Sb (III) by different adsorbents.

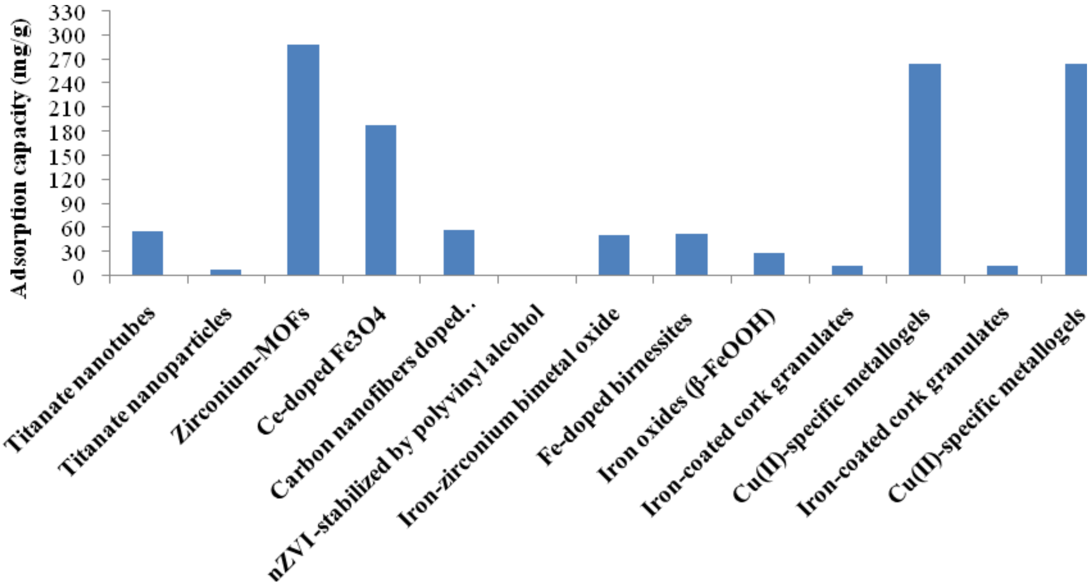

Fig. 3 Removal performance of Sb (V) by different adsorbents.

Table 1 Adsorption method for Sb removal from water and wastewater.

Type of adsorbent	Type of media	Time reaction	Species of Sb	pH	The best Model	Removal efficiency	Ref
Magnesium oxide	Wastewater	12 h	Sb (III)	7	Freundlich	140.1 mg/g	(Zhou et al., 2020)
Titanate nanotubes and Nanoparticles	Water	-	Sb (III) and Sb (V)	2	Langmuir	TiO_2 NTs: 250.0 and 56.3 mg/g, TiO_2 NPs: 12.0 and 8.6 mg/g	(Zhao et al., 2019)
Zirconium-Based Metal-Organic Frameworks	Water	10 h	Sb (III) and Sb (V)	2.3-9.5	-	136.97 mg/g and 287.88 mg/g	(Chmielewská et al., 2017)
Ce-doped Fe_3O_4	Aqueous solution	6 h	Sb (III) and Sb (V)	7	Langmuir	224.2 mg/g and 188.1 mg/g	(Qi et al., 2017)
Carbon nanofibers doped zirconium oxide	Aqueous solution	12 h	Sb (III) and Sb (V)	7	Langmuir	70.83 and 57.17 mg/g	(Luo et al., 2015)
Hydrochars and pyrochars	Aqueous solution	24 h	Sb (III)	3-6	Langmuir	Hydrochars: 2.24-3.98 mg/g and pyrochars: 4.44-16.28 mg/g	(Han et al., 2017)
nZVI-stabilized by	Artificial	48 h	Sb (III) and Sb (V)	7	Langmuir	6.99 and 1.65	(Zhao et al.,

polyvinyl alcohol	water					mg/g	2014)
ZVI/magnetite	Artificial water	2 h	Sb (III)	7	Langmuir	87.6 mg/g (97−99%)	(Li et al., 2018b)
Fe-doped birnessites	Artificial water	24 h	Sb (III) and Sb (V)	6	Langmuir	759 and 52.3 mg/g	(Lu et al., 2019)
Iron oxides (β-FeOOH)	Aqueous solution	24 h	Sb (III) and Sb (V)	9 and 4	Langmuir	34.09 and 29.22 mg/g	(Guo et al., 2014)
Iron-zirconium bimetal oxide	Water	1.5 h	Sb (V)	7	Langmuir	51 mg/g	(Li et al., 2012)
Iron-coated cork granulates	Water	24 h	Sb (III) and Sb (V)	6 and 3	Freundlich and Langmuir	5.8 and 12 mg/g	(Pintor et al., 2020)
Iron-modified attapulgite Nano-FeO(OH) modified clinoptilolite tuff	Aqueous solution	15 h	Sb(III)	<2.7	Freundlich	7.17 mg/g	(Chmielewská et al., 2017)
Cu (II)-specific metallogels	Water	40 min	Sb (III) and Sb (V)	4	Langmuir	102.4 and 264.1 mg/g	(Guo et al., 2018)
Fe (III)-treated humus sludge	Aqueous solutions	2 h	Sb (III)	2	Langmuir	9.433 mg/g	(Deng et al., 2018)
Raw pumice and Chitosan-modified Pumice	Aqueous solution	1.5 h	Sb (III)	5	Langmuir	44.8 and 88.9 mg/g	(Sari et al., 2017)

In a study, Qi et al. (2021) reported that the maximum adsorption capacity for Sb (V) removal from aqueous solution was 159.9 mg/g. They also stated that the common interfering ions of SO_4^{2-}, NO_3^-, CO_3^{2-}, PO_4^{3-} and SiO_3^{2-} exhibited an ignorable effect on Sb(V) elimination. Another study showed that the adsorption capacity of polymeric substances coating magnetic powders-supported nano zero-valent iron (nZVI@EPS@ Fe_3O_4) was 79.56 mg/g at pH=5, and 62% Sb (III) (79.56 mg/g) and 74% Sb (III) (91.78 mg/g) under aerobic and anaerobic conditions, respectively (Yang et al., 2020). A study by Yang He et al. showed that the highest adsorption capacity of nanocrystalline TiO_2 for Sb (III) and Sb (V) removal from contaminated environments was 588 and 333 mmol/kg, respectively. It also showed that Sb adsorption by TiO_2 was affected by pH, co-existing ions, humic acid (HA), etc (Yang et al., 2018). In another study, Wu et al. (2021b) reported that the efficiency of Sb (V) removal from water by franklinite-containing nano-FeZn composites was obtained to be 99.38% (543.9 mg/g) at initial Sb level of 9.173 mg/L, pH 6.45, adsorbent loading of 10 g/L, and the adsorption isotherms were fitted by the Langmuir model. In a study, Lapo et al. (2019) reported that the highest adsorption capacity of chitosan modified with iron (III) for Sb (III) removal from water was 98.68 mg/g at pH 6 and the adsorption process obeyed the Langmuir model. Another study by Wang et al. (2019b) indicated that the maximum adsorption capacity of hyper branched polyamide and sodium alginate microsphere was 195.7 mg/g for Sb (III) removal from wastewater (Sb (III) concentration range of 200-400 mg/L, and pH 5), and the adsorption process obeyed the Langmuir model. In a study, Bagherifam et al. (2022) showed that Fe (III)-modified montmorillonite could eliminate 95% of Sb (V) at concentration range of 0.2-1 mmol/L. Dong et al. (2021) found that the maximum adsorption capacity of diatomite coated with Fe-Mn oxides (DFMO) and pure Fe-Mn oxides (PFMO) was 10.7 mg/g and 2.8 mg/g for removal of Sb (V) from aqueous solution, respectively at pH of 3.5 and the adsorption isotherm obeyed Freundlich model. A study by Qi and Pichler (2016) demonstrated that Ferrihydrite could remove Sb (V) from aqueous phase by about 100% after 120 h at the Fe/Sb ratio of 500 and pH of 3.8. In a study by Wang et al. (2018), it was found that the maximum adsorption capacity of La-doped magnetic biochar for removal of Sb (V) from aqueous solution was 18.92 mg/g at pH of 7.0, and the coexisting Cl^- , SO_4^{2-} and NO_3^- exhibited ignorable interference on Sb (V) elimination. He et al. (2017) found that the efficiency removal of Sb (III) and Sb (V) from polluted water by amino modification of a zirconium metal–organic framework (MOF) was 61.8 mg/g and 105.4 mg/g, respectively within 20 min at pH 1.5-12. In a study by Wang et al. (2022), the adsorption capacity of Fe-Cu binary oxides for removal of Sb (V) from aqueous solution was found to be 94.3 mg/g at an initial level of 10 mg/L and pH 2-10, and the adsorption isotherms were fitted by the Langmuir and Freundlich models. A study by Wang et al. (2019a) found the adsorption capacity of Fe–Cu–Al trimetal oxide for Sb (V) removal from contaminated water to be 169.1 mg/g at an initial level of 0-60 mg/L and pH 7.0, and coexisting ions like chloride, sulfate, bicarbonate, silicate, phosphate, calcium and magnesium may affect Sb (V) adsorption. In their study, Bia et al. (2020) reported that Al_2O_3-supported Fe–Mn binary oxide nanoparticles (Fe–Mn@Al_2O_3) displayed the Sb (III) adsorption of 272.2 mg/g within 48 h at pH 7.0. They also showed that 92.8% of Sb (III) was sorbed by Fe–Mn@Al_2O_3, and 94.9% of sorbed Sb (III) was oxidized to produce Sb (V) and Mn (II) as quantitative removal mechanisms *reached* equilibrium. Moreover, they reported that 59.1% of the formed Sb (V) was adsorbed onto Fe-Mn@Al_2O_3 and 40.9% of Sb (V) was discharged into the solution (Bai et al., 2020). In a study, Xue et al. (2019) claimed that ZVI coupled with H_2O_2 at dosage of 0.5 g/L ZVI and 2 mM H_2O_2 at pH 3.0 could remove Sb from wastewater by 62.5%. A study by Guo et al. demonstrated that the Sb (V) elimination from water was above 90% with Fe_3O_4 magnetic nanoparticles coated with cationic surfactants at dosage of 0.4 g/L adsorbent, an initial level of 2-25 mg/L, and pH 3-5 (Meng et al., 2020). Zhou et al. (2020) found that the adsorption capacity of microbial extracellular polymeric substances-coated nano zero valent iron for Sb (V) from water was 202 mg after 2 h, pH 5.0, and adsorption isotherms were fitted by the Redlich Peterson model.

4.2 Biological and bioremediation techniques

Parameters affecting the remediation of Sb contamination included (1) plant and microbial communities (i.e. diversity, symbiotic interaction, enzymatic activity), (2) characteristics and weather (i.e. organic matter, water value, temperature, texture, pH, nutrients, redox potential), (3) contaminates and co-contaminates (i.e. concentration, toxicity, bioavailability, mass transfer), and (4) cost (i.e. pre-treatment, post-treatment, chemicals, amendments) (Bolan et al., 2022). Phytoremediation is an eco-friendly and inexpensive technique for Sb elimination from polluted media that can be used in situ or ex-situ. Phytoremediation is the use of plants to eliminate Sb bioaccumulation or decrease its toxicity. Currently, more than 60 Sb (III)-oxidizing strains have been isolated from Sb (III) contaminated media. These Sb (III)-oxidizing bacteria belong to 4 groups, including Actinobacteria 3%, Alphaproteobacteria 23%, Betaproteobacteria 25%, and Gamma proteobacteria 49%. Among these strains, Pseudomonas is the largest genus of Sb (III)-oxidizing bacteria (Li et al., 2022). In a plot scale study, Zhong et al. (2020) demonstrated that Sb is better adsorbed by plant roots in alkaline soil at pH value of 8.39 than acidic soil (pH 4.91). Another study by Cai et al. (2018) indicated that the adsorption capacity of *Bacillus subtilis* for Sb (V) removal from environment was 2.32 mg/g after 45 min, at 35°C, pH 5.0 and with biomass dose of 6.00 mg/L, and biosorption of Sb (III) obeyed the Freundlich model. A study by Uluozlu et al. (2010) showed that the removal of Sb (III) from aqueous solution with lichen (*Physcia tribacia*) was 81.1 mg/g within 30 min, at 20°C, pH 3.0 for the biomass dosage of 4 g/L. Vijayaraghavan and Balasubramanian (2011) found the adsorption capacity of algal (*Turbinaria conoides*) to be 18.1 mg/g for the removal of Sb (III) from aqueous solution within 45 min at an initial level of 10-100 mg/L, 296 K, pH 6.0; and biosorption of Sb (III) obeyed the Langmuir model. One study reported the oxidation of Sb (III) to Sb (V) using bacteria, such as hydrogenophaga, taeniospiralis, variovorax, and paradoxus, suggesting possible Sb (III)-dependent autotrophy (Terry et al., 2015). Wang et al. found that the Sb (III) oxidation by *Pseudomonas sp*. obeyed a zero-order kinetics (Li et al., 2022). In a study by Xi et al., the Sb (III) removal efficiency was reported to be 96.3% after 9 days of oxidation by sulfate-reducing bacteria (SBR). This study showed that 200-600 mg/L of Fe (III) increased the efficiency of total Sb removal. Moreover, it was found that the addition of Fe (II) can raise the hydrogenase activity of SRB and stabilize the pH of the SRB system (Lin et al., 2020). Another study indicated that bioremediation of Sb with sulfate reducing bacteria (SBR) reduced Sb (V) to Sb (III) at pH 7 and 9. This technique is called a cost-effective biological process to remove Sb (V) from sulfate-laden wastewaters (Wang et al., 2013). Zhao et al. reported the Sb retention in roots of cork oak (*Quercus variabilis* Bl.) to be 1623.39 mg/g (Zhao et al., 2015). Bech et al. (2012) reported the accumulation of Sb using various plant species (*Poa annua* L., *Echium vulgare* (L.), *Sonchus asper* (L.) Hill, *Barbera verna* (Mill.) Asch.) to be between 1.21 mg/ kg and 4.9 mg/kg. In a study, *Pteris vittata* L. (PV) plant exhibited the highest values of Sb accumulation, and the range of Sb concentration in roots, stems, and leaves was 1.2-140.1, 1.0-29.7, 6.0-73.4 mg/kg, and 0.49-546.7, 0.19-3.1, and 1.6-129.9 mg/kg for PV and *Miscanthus floridulus* (MF) plant, respectively (Ran et al., 2020). In a study, Ren et al. (2019) reported the total Sb in deposits from the two regions to be 3.50 mg/kg and 3.21 mg/kg in the algae-and macrophyte-dominated regions, respectively, this resulted from the oxidation of Sb (III) to Sb (V) by Mn and Fe oxides in both zones under aerobic conditions. One study reported that the biogenic Mn oxides (BMO) is able to oxidize Sb (III) to Sb (V) and to further adsorb Sb (V) to its surface (Bai et al., 2017). In a study, Xu et al. (2022) reported the removal of Sb (III) and Sb (V) from aqueous solution with Nano–silica and biogenic iron (oxyhydr) oxides composites (BS–Fe) to be 102.10 mg/g and 337.31 mg/g, respectively, and the adsorption data were fitted by the Langmuir and Temkin models at 24 h and pH 2 to 12.

Table 2 Biological and bioremediation method for Sb removal from water and wastewater.

Type of adsorbent	Type of media	Sb Species	pH	Initial concentration	Removal efficiency	Ref
Phytoremediation (*Pteris cretica* L)	Aqueous media	Sb total	7.8	10 and 20 mg/L	1516.5 mg/ kg	(Feng et al., 2011)
Phytoremediation (*Pteris cretica* Albo-Lineata’, *Pteris fauriei*, *Humata tyermanii* Moore, and *Pteris ensiformis* Burm	Aqueous media	Sb total	6.8	20 mg/L	6405 mg/ kg	(Feng et al., 2015)
Phytoremediation (*Pteris vittata*)	Aqueous media	Sb total	-	5 to 16 mg/g	230 mg/ kg	(Müller et al., 2013)
Biosorption (Cyanobacteria Microcystis)	Surface water	Sb (V)	2.5-3.0	10 mg/L	nearly 90%	(Sun et al., 2014)
Biosorption (*Geotrichum fragrans*)	Aqueous media	Sb (V)	3.4	20 mg/L	99.8%	(Wan et al., 2014)
Biosorption (seaweeds)	Aqueous media	Sb (III)	7	25 mg/L	4mg/g	(Ungureanu et al., 2017)

Fe (III) modified *Proteus cibarius*	Aqueous media	Sb (III)	6.0	27.70 mg/L	30.612 mg/g (97.60%)	(Zeng et al., 2021)
Bioreactor (Ferrovum, Thiomonas, Gallionella, and Leptospirillum)	Mine waters	Sb (III) and Sb total	-	629-20, 629 mg/kg	80% and 90%	(Sun et al., 2016)
Bioreduction (*Dechloromonas* sp. AR-2 and *Propionivibrio* sp. R-3)	Waste water	Sb (V)	6-7	5 mM	Over 74%	(Yang et al., 2021)

4.3 Coagulation and flocculation

Soluble Sb (III)/(V) is changed into insoluble products by coagulants and sedimented through adsorption, inter-particle bridging, precipitation and co-precipitation, then is eliminated by sedimentation or filtration. Like adsorption, this technique is advantageous due to its simple operation, being inexpensive, and easy handling coagulants (ferric chloride, poly aluminum chloride, poly ferric sulfate and ferrous sulfate). Wu et al. (2010) reported that the Sb (V) elimination illustrated a considerable and continual reduction as the competing ions (e.g., bicarbonate, sulfate, phosphate, and HA) increase. Besides, Sb (III) was less affected by the interfering constituents like phosphate and HA. A study by Kang et al. (2003) indicated that the elimination of both Sb (III) and Sb (V) by coagulation with ferric chloride was much higher than that of polyaluminum chloride, and removal efficiency was about 67% using a high coagulant dosage of 10.5 mg of Fe/L at optimal pH of 5.0. A study by Liu et al. indicated that the highest Sb (V) removal efficiency reached up to approximately 95.0% with coexistence of 100 mg/L dispersive dye at pH 3, with a dose of 200 mg/L (Tang et al., 2020). In another study, Liu et al. (2019) found the Sb (V) removal from wastewater to be 93.8% at pH 6 using aerated PFS/$FeSO_4$ coagulation. In a study by Zhu et al. (2011), the removal efficiency of Sb from wastewater using electrocoagulation was reported to be 96-100% after 60 min at pH 4-6 and current density of 166.67 A/m^2. Song et al. (2015) used electrocoagulation with hybrid Fe–Al electrodes for Sb removal from surface water and obtained the Sb (V) removal percentage to be more than 99% after 89.17 min, at initial concentration of 521.3 µg/L, pH 5.24, and 2.58 mA/cm^2 current density. A study by Jiaxing et al. (2014) indicated that the amount of Sb (III) and Sb (V) was reduced to 5.0 and 28.1 µg/L, respectively after 30 min using electrocoagulation method to treat Sb pollutants in water. In a study by Mitrakas et al. (2018), the adsorption capacity of iron coagulants was 4.7 Sb (III)/Fe (III) µg/mg and 0.45 Sb (III)/Fe (II) µg/mg for Sb (III) and Sb (V), respectively at pH 7. Using polyethylene terephthalate resin and $FeCl_3 \cdot 6H_2O$, $FeSO_4 \cdot 7H_2O$, $AlCl_3 \cdot 6H_2O$, and $TiCl_4$ salts as coagulants, Vengris et al. obtained Sb removal from wastewater to be about 98% at initial concentration of 1200 mg/L and pH of 4-9 (Mitrakas et al., 2018).

Table 3 Coagulation and flocculation method for Sb removal from water and waste water.

Type of method	Type of media	Species of Sb	pH	Initial concentration	Removal efficiency	Ref
Hybrid coagulation-flocculation-ultrafiltration	Surface water	Sb (III)	7.1-9.0	30.0 to 158.0 µg/L	over 90%	(Du et al., 2014)
Coagulation-flocculation-sedimentation	Aqueous solution	Sb (III) and Sb (V)	4–6	100 and 250 µg/L	About 90% and 99.7%	(Guo et al., 2018)
Coagulation(ferric chloride)	Drinking water	Sb (III) and Sb (V)	7	101 and 98.4 µg/L	86 to 97% and 92 to 96%	(Wu et al., 2010)

Coagulation (ferric chloride)	Synthetic water	Sb (III) and Sb (V)	7-8	1 mg/L	71.02 %and 67.03%	(Inam et al., 2019)
Coagulation (ferric chloride)	Aqueous solution	Sb (III) and Sb (V)	7	0.1 to 0.9 mg/L	92.7%	(Inam et al., 2018)
Coagulation (ferric chloride)	Aqueous media	Sb (III) and Sb (V)	4 to7	10 mg/L	90–100%	(Inam et al., 2018)
Coagulation (polyferric sulfate and ferrous sulfate)	Wastewater	Sb (V)	5 to 6	500 μg/L	93.8%	(Liu et al., 2019)

4.4 Membrane filtration

Membranes play an important role in chemical techniques and are used in a wide range of applications, especially in the advanced treatment of drinking water. Among the membrane techniques used for Sb removal from water and wastewater are ultrafiltration (UF), RO, nanofiltration, and electrodialysis. Limitations of these processes include the need for power use, financial cost, and the need for operation factors optimization. One study reported that RO could remove more than 90% of Sb from wastewater at a pilot-scale study. UF was also unsuccessful for the elimination of Sb (Wu et al., 2021a). A study by Kang et al. (2000) indicated that RO could remove approximately 60.2% of Sb (III) at pH=3-10. It also showed the superiority of the effect of RO membrane on Sb elimination as compared with arsenic (As). Moreover, the study indicated that the retention effect of RO membrane on Sb(V) is better than on Sb (III). This superiority can be attributed to the electrical properties of the present species of Sb in water with various valence states (Kang et al., 2000). Ran et al. (2020) reported the highest value of 97.29% for Sb (V) removal from water using coagulation-floc preloaded ultrafiltration (CFPLU) treatment and stated that Sb (V) concentrations of CFPLU under all solid levels are below 5 μg/L.

Table 4 Membrane filtration method for Sb removal from water and wastewater.

Type of method	Type of media	Species of Sb	pH	Initial concentration	Removal efficiency	Ref
Ultrafiltration membrane	Water	Sb (V)	6.0	5.4 μg/L	92.8%	(Ma et al., 2017)
Forward osmosis	Wastewater	Sb (III)	3 and 7	500 μg/L	About 99.7%	(Song et al., 2020)
Membrane Bioreactor	Wastewater	Sb (V)	-	100 μg/L	About 95%	(Komesli, 2014)
Polysaccharide functionalized hybrid membrane	Aqueous solution	Sb (III) and Sb (V)	3	2 to 60 mg/L	16.5 and 13.6 mg/g	(Zeng et al., 2021)

4.5 Photooxidation

The conversion of Sb (III) to Sb (V) may be mediated by chemical processes, such as ferrous iron-induced •OH, Fe oxyhydroxides, oxidized by $^{\bullet}Cl_2{}^-$ and $^{\bullet}OH$ radicals, and irradiation of light. A study by Kong et al. indicated that in acidic conditions (pH 1–3), •OH and $\bullet Cl_2$ generated by the photocatalysis of $FeOH_2{}^+$ and $FeCl_2{}^+$ are the main oxidants of Sb (III) (Kong et al., 2016). One study showed that antimony trioxide (Sb_2O_3) was oxidized to Sb (III) at pH 3.0-9.0 under the simulated ultraviolet ray (Hu et al., 2014). Another study by Wu et al. showed that the Sb (III) was quickly oxidized to Sb (V) in the presence of various phenolic acids in neutral and alkaline states, and the highest oxidation of Sb (III) was observed at pH 9, and kinetic data were fitted by the pseudo-first-order(Wu et al., 2019).In a study, Quentel et al. (2006) reported the pH dependency of Sb (III) oxidation with iodate and stated that no measurable oxidation was observed below pH 9. A study by Kong et al. (2015) indicated that the Sb (III) was oxidized to Sb (V) with pyrite-induced hydroxyl radical (·OH) and hydrogen peroxide (H_2O_2), and the reaction improved as the pH increased. One study reported the oxidation of Sb (III) via atmospheric oxygen at 25^oC and considerable oxidation within 4-420 days at pH 12.9 (Multani et al., 2016). Buschmann et al. (2005) reported photooxidation of Sb (III) to Sb (V) in the presence of HA at pH 2-11 using UV-A and visible light, and stated that the oxidation rates can be well predicted based on dissolved organic carbon.

Table 5 Photooxidation method for Sb removal from water and wastewater.

Type of method	Type of media	Species of Sb	pH	Initial concentration	Removal efficiency	Ref
Photo-induced oxidation	Aqueous media	Sb (III) and Sb (V)	5 and 3	50 μmol/ L	100% and 97%	(Sun et al., 2014)
Photooxidation (organic Fe (III) complexes)	Aqueous media	Sb (III)	3.0 and 6.2	20 μM	68.6% and 36.6%	(Kong and He, 2016)

4.6 Electrochemical method

Electrochemical techniques are essential and enabling disciplines in environmental treatment, such as the removal of pollutants, water sterilization and disinfection, and recycling of pollutants. One study reported the Sb removal from acidic aqueous solution with an electrochemical method to be 1500 mg/L-3500 mg/L (Bergmann and Koparal, 2007). In another study, Bergmanna and Koparal (2011) investigated the Sb removal using electrochemical deposition process with copper and graphite electrodes and found that Sb concentration reduced from 5 mg/L to 0.15 mg/L by changing the electrode material, electrolyte type, and current of cell. Staicu et al. (2015) reported the Sb removal from water using electrochemical treatment with Al and Fe electrodes to be 54% and 93%, respectively, at initial concentration of 310 mg/L at pH7. In a study by Awe et al. (2013), the Sb (III) removal from aqueous solution using an electrowinning method was reported to be 89% at initial concentration of 25-35 g/L at pH4. Yilmaz et al. (2018) used electrocoagulation process with aluminum and iron electrodes for chemical oxygen demand (COD) removal from vinegar industry wastewater and obtained a removal efficiency of 90.91% at pH 4, 20.00 mA/cm^2 current density; and 93.60% at pH 9, 22.50 mA/cm^2 current density, respectively.

Table 6 Electrochemical treatment for Sb removal from water and wastewater.

Type of method	Type of media	Species of Sb	pH	Initial level	Removal efficiency	Ref
Electrochemical	Wastewater	Sb (V)	4-7	10-120 mg/L	99%	(Cao et al., 2017)
Electrocoagulation	Wastewater	Sb Total	4 to 6	1 mg/L	96%	(Zhu et al., 2011)
Electrodialytic	Environment	Sb Total	Below 4	-	Below 20%	(Pedersen et al., 2018)

4.7 Other technologies

Other treatment techniques that can be used for Sb removal from water and wastewater include ion-exchange, oxidation process, manganese sand filter, and Standard Oil of Ohio Company (SOHIO) acrylonitrile process. One study reported that the XAD-8 ion-exchange resin can eliminate Sb because the ion exchange resin has a strong exchange capacity for Sb (III) and Sb (V) (Gao et al., 2015). The study of the ion exchange behavior of Sb on anion exchange resins showed that the absorbability for Sb (III) was higher than for Sb (V), and it was further complicated to elute Sb (III) as compared with Sb (V) (Guin et al., 1998). A study by Wang et al. indicated that 95% of the Sb (III) oxidation process was done by the particles (i.e. macroparticles 38%, colloids 23%, and dissolved substances 34%) in natural water. The mechanism of Sb (III) oxidation and adsorption include the formation of $O2^{\bullet -}$, •OH, and dissolved organic matter (DOM) with different particle size (Wang et al., 2020b). Liu et al. found the efficiency of Sb removal from wastewater using Fe_3O_4 combined with manganese sand filter to be 92% after 20 min at pH 3. Findings of their study showed that the addition of Fe_3O_4 improved the elimination of Sb in this process (Tang et al., 2020). A study by Foste et al. (2019) found the elimination efficiency to be 99.6% and 99.4% for Sb (III) and Sb (V) removal from wastewater, respectively at initial concentration of 50 mg/L pH5 with ferric chloride dose of 460 and 470 mg/L using Standard Oil of Ohio company (SOHIO) acrylonitrile process.

Table 7 Other techniques used for Sb removal from water and wastewater.

Type of method	Type of media	Species of Sb	pH	Initial concentration	Removal efficiency	Ref
Ion-exchange	Wastewater	Sb (III) Sb (V)	-	200 mg/L	Sb (III) 97%, Sb (V) 16%	(Riveros, 2010)
Ion–exchange(anion exchange)	aqueous solution	Sb (V)	9	0.5 mol/L	Above 90%	(Kameda et al., 2012)
Hydrated ferric oxides using a polymeric anion exchanger and calcite	Synthetic water	Sb (V)	3 to 9	5 to 70 mg/L	About 84%	(Miao et al., 2014)
Organic ligand-induced	Aqueous media	Sb_2O_3	3.7,6.6, and 8.6	-	97%	(Cao et al., 2017)

4.8 Antimony (Sb) from global perspectives

According to findings of this study in ground waters, the highest Sb concentration of 13.5-33.2 μg/L was achieved in Nigeria and the lowest concentration of 0.71 μg/L was found in Poland (Poznan) (Table 9), while in surface waters, the highest Sb concentration of 100-7000 μg/L was found in China (Xikuangshan), and the lowest level of 0.029-0.736 μg/L in China (Yangtze River) (Table 9). The WHO and USEPA guideline value for Sb was set as 5, and 6 μg/L, respectively (Table 8). However, the levels mentioned in the literature exhibit a very broad range from 0.00 to 7000 μg/L. One study reported that a greater percentage (>70%) of the sampled wells recorded the mean Sb values a little higher than the safe limit of 20 μg/L for potable water set by the WHO (Etim, 2017). One study indicated that the general mean Sb value in groundwater within the metropolis was 24.9±26.2 μg/L (Etim, 2017).

Table 8 International Antimony guidance levels for potable water.

Country/Organization	Concentration (μg/L)	Ref
World Health Organization (WHO)	5	(Inam et al., 2019)
United States Environmental Protection Agency (USEPA)	6	(Luo et al., 2015)
European Environment Agency	6	(Luo et al., 2015)
European Union (EU)	10	(Du et al., 2014)
Japan	Less than 2	(Zhao et al., 2019)
China	5	(Amarasiriwardena and Wu, 2011)
Korea	20	(Inam et al., 2019)
Australian	3	(NHMRC, 2011)
Iran	20	(Iran, 2010)

Table 9 Antimony (Sb) concentrations in different water from several countries.

Regions/countries	Water sources	Antimony concentration (μg/L)	Ref
China (Hunan Province)	Surface water (river)	2.0-6384	(Tang et al., 2020)
China (Xikuangshan)	Surface water	100-7000	(Qi et al., 2017)
China (Yangtze River)	Surface water	0.029-0.736	(Wu et al., 2011)
China (Bohai Bay River)	Surface water	0.386-1.075	(Duan et al., 2010)
Slovakia	Surface water	< 1-2150	(Hiller et al., 2012)

Slovakia	Surface water	1000	(Inam et al., 2019)
USA (Alaska)	Surface water	239	(Inam et al., 2019)
Turkey (Balcova)	Surface water	0.00-24	(Aksoy et al., 2009)
Italy	Surface water	0.5-148	(Armiento et al., 2017)
Mexico	Surface water	1.2-220.60	(Baeza et al., 2010)
Iran (Takab)	Surface water	0.65-8.95	(Sharifi et al., 2016)
Norway	Groundwater	0.002-8.00	(Li et al., 2018a)
Turkey (Balcova)	Groundwater	0.06-26	(Aksoy et al., 2009)
Finland	Groundwater	0.02-0.82	(Lahermo et al., 2002)
Nigeria	Groundwater	13.5-33.2	(Etim, 2017)
Ethiopia	Groundwater	0.002-1.780	(Reimann et al., 2003)
Poland (Poznan)	Groundwater	0.71	(Niedzielski and Siepak, 2005)
Turkey (Balcova)	Geothermal water	0.7-170	(Aksoy et al., 2009)
France	Mining water	0.0067-0.156	(Elbaz-Poulichet et al., 2020)
North Pacific	Marine waters	0.09-0.14	(Morel and Price, 2003)
North Atlantic	Marine waters	0.21	(Morel and Price, 2003)
Western Atlantic ocean	Marine waters	0.13	(Cutter et al., 2001)

Total background concentrations of dissolved Sb in groundwater have been explained in the range of 0.010-1.5 µg/L (Mitrakas et al., 2018), while anthropogenic and geothermal sources are accountable for much higher concentrations in ranges of 0.7-170 µg/L and 0.06-26 µg/L, respectively (Aksoy et al., 2009). A study by Alderton et al. (2014) indicated that surface waters contained a higher level of Sb than the groundwater (2.1-0.6 µg/L, medians). One study reported the mean Sb level in the rivers worldwide to be 1 mg/L (Ungureanu et al., 2015). The Sb concentrations in numerous domestic wells of Slovakia exceeded the Sb potable water limit of the WHO guideline value by as much as 25 times. The Sb levels in clean water and seawater are below 1 µg/L and 0.2 µg/L, respectively, and up to 100 µg/L in the proximity of an anthropogenically polluted water (Bolan et al., 2022).

5 Conclusions

Hazardous Sb pollution of water and wastewater is one of the most important environmental challenges in a few countries. Different techniques have been used and proposed to eliminate Sb from aqueous phase. Considering legal limits and toxic effects, the elimination of these metalloids from water and wastewater (due to anthropogenic or natural causes) is compulsive. Currently, RO, photooxidation, electrodeposition, precipitation/coagulation and membrane filtration, electrocoagulation, ion exchange, biosorption, and adsorption technology are used in varying degrees. It is concluded from this study that much present research is principally focused on adsorption and biological/bioremediation techniques. Adsorption by low-cost adsorbents and biosorbents is recognized as an effective and economic technique for treatment of water and wastewater with low concentration of Sb. Techniques such as biosorption, RO, photooxidation, and coagulation treatment show the best results among all (>99%). It is not possible to select the best method because each one has its own strengths and limitations, and the content of the application has to be considered. It is thus essential to fill the gap between theoretical research and real application and develop the hybrid and effective techniques for simultaneous elimination of high or low concentrations of Sb from water and wastewater. Some challenges in application of these techniques include energy consumption, generation of toxic sludge, using high coagulant values, financial cost, generation of secondary pollution, being ineffective for low concentrations of Sb, sensitivity to water and wastewater quality, clogging and fouling of membranes, regeneration and recycling, and reuse of adsorbents. These challenges should be considered in more studies to find more reliable and sustainable solutions for them. In sum, most studies at present only discuss the effect of a single factor or lack of in-depth studies. Hence further evaluation is needed to describe all factors' effects, mechanisms, and techniques.

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