



# REMOVAL OF ARSENIC AND PHARMACEUTICAL COMPOUNDS FROM DRINKING WATER: A META-ANALYSIS

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**Abstract-** Arsenic being a global contaminant and highly toxic metalloid has gained much attention for many years. Intending to provide potable water to millions of people around the globe, the research community has practiced intensive efforts to remove arsenic. This review is focused on the possible treatment methods which can be deployed to curb arsenic contamination. Apart from discussing arsenic, this review also explored the occurrence and removal strategies of pharmaceutical contaminants. Unlike conventional contaminants, the problem associated with pharmaceutical contaminants is not their presence in abundance but rather very minute concentrations. Nearly undetectable concentration of pharmaceutical compounds, and their unpredictable impacts on the environment and humans are fundamental concerns for the research community. It has been found that conventional treatment technologies are insufficient for the remediation of pharmaceutically contaminated environmental segments; therefore advanced oxidation processes, membrane technologies, adsorption processes, and various integrated approaches have been evaluated in this paper. This review provides an insight into the advances being made till now in removing such contaminants from water.

**Keywords-** Arsenic, Emerging contaminants, Meta-analysis, Pharmaceutical compounds, Water treatment.

## 1 Introduction

Arsenic (As) is a highly lethal and cancer-causing metalloid that creates approximately 0.00005% Earth's crust and distributes in an extensive range via groundwater and surface water systems[1]. Arsenic can be found in inorganic and organic forms, generally two forms of arsenic found in aqueous environment including arsenite "As(III)" and arsenate "As(V)" [2]. It has an atomic weight of 74.9 g.mol<sup>-1</sup> and a specific gravity of 5.73 g.cm<sup>-3</sup> with boiling and melting points of 614°C and 817°C, respectively. In terms of mobility, the As(III) is highly mobile (25–65 times) in the natural environment as compared to As(V), predominantly due to its electro neutrality[3]. It exists as a deformable crystal-like solid material with a silver grey color. Moreover, it is tasteless, transparent, odorless, and difficult to detect. Since arsenic occurs naturally in geologic environments, therefore, groundwater pollution may arise because of mining, agricultural, and industrial practices. It is also known for its toxicity to humans and marine species [4, 5]. Researchers identified arsenic pollution as a 21st century disaster. Before the year 1993, the World Health Organization (WHO) endorsed 0.05 mg/L as a threshold limit for arsenic in drinking water. Later, the Environmental Protection Agency (EPA) approved a new lower limit of 0.01mg/L in the year 2001. Keeping in view the immediate toxicity and long-term consequences of arsenic, ranging from mild skin disease to cancer, the World Health Organization (WHO) further decreased its nontoxic limit to 10µg/L due to its hazardous nature. For instance, consumption of inorganic arsenic-contaminated rice and prolonged exposure to water containing its high concentrations (i.e., >100 mg/L) cause lung ,bladder cancer and other organ failure [6, 7].

Pakistan is at the 80th position out of 122 countries in the world, which are facing severe arsenic contamination in water. In Pakistan, 3% of the population of Punjab is exposed to a concentration of greater than 50 g/L of arsenic and 20% of Punjab's population is exposed to greater than 10 g/L concentration of arsenic. Similar to Punjab, situation is worse in



Sindh, where 16% of the population is said to be exposed to greater than 50 g/L and 36% to greater than 10 g/L concentration of arsenic [8].

Apart from Arsenic, the presence of emerging micro-pollutants (EMPs) found in water bodies is also a matter of rising concern at the global level [9]. Major sources of these pollutants include industrial chemicals, illicit drugs, pharmaceutical and personal care products (PPCPs), perfluorinated compounds, and flame retardants [10]. Their presence has been identified in various natural environments including surface and groundwater sources along with water and wastewater treatment facilities [11]. However, their monitoring is globally out of reach as there is a scarcity of data about the quantity and occurrence of EMPs in water [12].

Among various EMPs, pharmaceutical compounds are getting more attention due to their persistence and toxicity [13, 14]. Pharmaceuticals are a vast range of chemicals used in medicine for both humans and animals [15]. Although their presence in water bodies has been identified for decades, however, their concentration in the natural environment has just started to be recognized as potentially dangerous and unsafe for the ecosystems [16]. Even the small concentrations of pharmaceutical compounds in drinking water supplies can cause harmful effects on the health of humans. The existence of pharmaceuticals in water comes from hospital waste, therapeutic drugs, and pharmaceutical industry waste. Due to the increase in human pharmaceutical consumption, various pharmaceutically active compounds (PACs) are disposed into the environment each day [17]. This increasing usage has raised queries related to their harmful risks to the environment, quality of water, and human health [18]. The absence of discharge standards and relevant regulations is a major reason behind these issues. Even after the conventional treatment, many of the PACs remain in the effluent and be part of the environment [19].

Therefore, the focus of this review is on the potential treatment technologies that can be utilized to remove both arsenic and pharmaceutical contaminants from water. Contrary to ordinary pollutants, these micro-pollutants are problematic when they are present at extremely low levels. The research community has shown serious concerns about the nearly undetectable concentration of pharmaceutical contaminants and their unpredictable effects on the environment and human beings. Since conventional treatment methods are ineffective for the removal of such contaminants, therefore, an attempt has been made in this review to evaluate various advanced treatment methods (e.g., advanced oxidation processes, membrane technologies, adsorption processes, and their integrated schemes) for the effective treatment of such toxic contaminants.

## **2 Treatment Methods used for the Removal of Arsenic**

The status of arsenic as a global contaminant has not faded yet. According to the United States Geological Survey (USGS), developed and developing countries are exposed to the threat posed by arsenic and emerging contaminants [20]. Currently, their sustainable removal is a challenge for the scientific community. To overcome this issue, extensive research has been conducted on arsenic removal. The methods used for this purpose include adsorption, ion exchange, chemical precipitation, phytoremediation, electro-coagulation, phytobial remediation, and electro-kinetic method [21]. These methods along with their strengths and limitations have been described in the following sections.

### **2.1 Removal of Arsenic by using the adsorption method**

Adsorption is an efficient and cost-effective arsenic removal method, which is directly applicable in developing regions having a lack of qualified staff and fluctuations in electrical supply. Recently adsorption has provided more than 95% efficiency for the removal of arsenite and arsenate [22]. This process is primarily determined by the separation process using the Van der Waals concept and the electrostatic attraction forces. The efficiency of this method depends upon various process parameters including pH, exposure time, presence of other reactive species, adsorbent concentration, original arsenic absorption, and temperature. Various adsorbents used for arsenic removal include zero-valent iron, metal oxides coated in sand, iron oxide-based sorbents engineered biochar (80% removal), granular adsorbents (35% removal), clinoptilolite-rich zeolitic tuff, activated carbon (20-40% removal), and activated alumina (>85% removal) [23-25].

### **2.2 Removal of Arsenic by using the Ion-exchange method**

Ion exchange is a physicochemical treatment process. Ions electrostatically maintained on the hard surface of ion-exchange resin solution are swapped from solution with the ion of a similar charge [26]. The United States Environmental Protection Agency (USEPA) also suggested specific ion exchange resins for arsenic subtraction [27]. The process parameters influencing the efficiency of this method include the concentration of arsenic, type of resin, high sulfate salts, competing ions, and total dissolved solids (TDS). Previously, Ortega, et al. [28] reported 18.9% arsenic removal efficiency by using the ion-exchange method.



### 2.3 Removal of Arsenic by using the chemical precipitation method

Chemical precipitation uses mixtures such as sulfides, calcium, magnesium, and ferric salts for treatment. These reagents aid in the removal of arsenic by reducing dissolved arsenic to molecules with limited solubility compounds [29]. Previously, two-stage nanofiltration and chemical precipitation were used to treat gold excavation wastewater for arsenic and calcium removal [30]. Similarly, removal efficiencies of 98.5%, 96.3%, and 26.5% were reported by using lead hydroxide, lead nitrate, and lead oxide, respectively. Since this process produces toxic sludge, therefore pre-oxidation is necessary which results in the production of hazardous disinfection byproducts. To overcome this issue, sequential oxidation, chemical treatment, sedimentation, and filtration all are required for treatment.

### 2.4 Removal of Arsenic by using the phytoremediation method

This process utilizes plants for the removal of contaminants with minimum fertilizer input, thereby making it socially acceptable and environmentally sustainable [31]. These plants generally have deep roots, high tolerance, fast development, and are capable to absorb arsenic from contaminated soil [31, 32]. For this purpose, some plant species use direct absorption for aerial metal buildup, whereas, other plant species transfer metals via transporter proteins [33]. Studies also suggest combining nanomaterial with plants and microbes to enhance conventional bioremediation methods [34]. For instance, Yang, et al. [35] reported an arsenic accumulation of up to 171% by inoculating the process with *Cupriavidus Basilensis* bacteria. However, the non-specific presence can harm the surrounding environment. Since the removal efficiency is dependent on the type of macrophyte, therefore, further research is required to better understand this process [34].

### 2.5 Removal of Arsenic by using the electrocoagulation method

It is one of the most prominent methods for the removal of arsenic from water. This process has a high removal efficiency of 93-99.9% for arsenic removal [36], however, its limitations include (i) complexity, (ii) the electrodes get passive during the removal process, (iii) high power consumption [37], (iv) high cost, and (v) sludge production. For instance, the cost of arsenic removal through this process is 1.8 times of a simple coagulation process due to high energy consumption [38]. Despite various attempts have been made to overcome the aforementioned limitations, however, further research is required for its optimization [39].

### 2.6 Removal of Arsenic by using the phytobial remediation method

It is an inexpensive and environmentally friendly method. The microbe-assisted plant remediation has been widely studied for the degradation of arsenic in rhizophores [40]. Although this process provides 99.9% arsenic removal efficiency [41], however, it is time-consuming due to slow plant growth and the sensitivity of plant species to arsenic concentration. The use of bacteria for the degradation of arsenic through genes is complex and is in the research phase for arsenic removal [42, 43].

### 2.7 Removal of Arsenic by using the electro-kinetic method

This method has been used to remove arsenic from soil and dissolved heavy metals from tainted groundwater [44]. Since this treatment method cannot remove arsenic in its dissolved phase, therefore, using an integrated treatment would be an environmentally friendly, technically efficient, and economically viable solution [45]. This aspect demands further research to investigate the integration of the electro-kinetic method with other treatment techniques. A summary of arsenic removal efficiencies achieved by using above mentioned methods has been provided in Table 1 [46].

Table 1: Arsenic removal efficiencies achieved by using different treatment methods.

Method	Arsenic Removal Efficiency
Adsorption	95%
Ion Exchange	18.9%
Chemical Precipitation	98.5% by using lead hydroxide 96.3% by using lead nitrate 88% by using A. Pinnata
Phytoremediation	83% by using L. Minor 78% by using H. Verticillate 85.5% by using their mixture
Electrocoagulation	93-99.9%
Phytobial remediation	99.9%



Table 1 is an illustration of different arsenic removal efficiencies that have been achieved by using various treatment methods. Among them, the adsorption process has been used extensively due to the easy availability of adsorbent materials, simple operation, and cost-effectiveness.

### 3 Treatment methods used for the removal of pharmaceutical compounds

The use of physical, chemical, and biological treatment processes in conventional treatment plants can eliminate a plethora of contaminants, however, they are not specially designed to eliminate new pollutants like antibiotics to safe permissible levels [47]. Therefore, relying solely on conventional treatment methods would not fulfill the task of removing emerging contaminants (ECs). Various researchers examined the possibility of removing emerging contaminants (ECs) using these methods, but overall results remained unsatisfactory [48-50]. For instance, Sun, et al. [51] claimed the inadequacy of conventional methods for the subtraction of polyfluoroalkyls and perfluoroalkyls. Similarly, He, et al. [52] described the incomplete elimination of tetracycline using the coagulation-flocculation method by utilizing polyferric sulfate, polyaluminium chloride, and polyaluminium sulfate as coagulants. Likewise, Petrovic, et al. [53] achieved only 7% removal efficiency of ECs by using the coagulation-flocculation followed by rapid sand filtration. To overcome these limitations, several advanced treatment methods have been developed by previous researchers including adsorption, membrane technologies, and advanced oxidation processes (e.g., Fenton/Fenton-like methods) as described below.

#### 3.1 Removal of pharmaceuticals by using adsorption

In this process, mass is transferred between the liquid-solid phase, gas-solid phase, liquid-liquid phase, or gas-liquid phase [54]. Contaminants adsorb on the surface of the adsorbent via intermolecular forces such as electrostatic attraction, hydrophobic interfaces and Van der Waals forces. Previously, different types of adsorbent materials have been used for the removal of ECs including activated carbon [55], graphene oxides [56], biochar [57], chitosan [58], and iron oxides [59]. The removal efficiency of these adsorbents depends on the type of targeted ECs. For instance, activated carbon gave complete removal of ciprofloxacin [60], however, the results were not satisfactory for the removal of paracetamol and tetracycline [61]. Similarly, biochar has also been used as an adsorbent for the removal of ECs [62], where thermochemical conditions maintained during the synthesis of biochar greatly affect the adsorption efficiency [63].

Likewise, Azqhandi, et al. [64] observed the complete removal of ciprofloxacin by using montmorillonite clay. Furthermore, alum-based adsorbents and zeolite have also been utilized for the removal of such compounds from an aqueous medium. The detail of ECs removed by various adsorbent materials has been provided in Table 2 [65].

Table 2: Adsorption capacities of different adsorbent materials for the removal of various emerging contaminants.

Emerging contaminants	Adsorbent type	Adsorption capacity (mg. g <sup>-1</sup> )
Perfluoro octane sulfonate	Polyaniline nanotubes	1651
Methyl paraben	Activated carbon chitosan composite	85.90
Propyl Paraben	Activated carbon chitosan composite	90.0
Perfluoro octane sulfonate	Bentonite	99.90
Phenanthrene	Bentonite	4.05
Perfluoro octanoate	Polyaniline nanotubes	1100
Estriol	Graphene	200
Imidacloprid	Peanut husk	6.53
17b-estradiol	Modified activated carbon cloths	12.66
17a-ethinylestradiol	Modified activated carbon cloths	11.11
P-chloro-m-xylenol	Powder activated carbon	1.3
Bisphenol-a	Bagasse-b-cyclodextrin polymer	121
Atenolol	Oxidized activated carbon	100.6
Diazinon	Fe <sub>3</sub> O <sub>4</sub> -guar gum-montmorillonite	80.0
Diazinon	Fe <sub>3</sub> O <sub>4</sub> -guar gum	47.17
Paraquat	Modified zeolite	166.71
Triclosan	High-silica zeolites	378
Metalaxyl	Natural clay	0.09
Atrazine	Graphene nanosheets oxide	23.84



Emerging contaminants	Adsorbent type	Adsorption capacity (mg. g <sup>-1</sup> )
Tricyclazole	Natural clay	0.08
Tetracycline	Biochar	58.80
4-chlorophenoxyacetic acid	Activated carbon	0.002
Clofibric acid	Activated carbon	0.002
Acetaminophen	Double-oxidized graphene oxide	704
Carbamazepine	Powdered activated carbon	0.04
Diclofenac	Powdered activated carbon	0.04
Sulfamethazine	Biochar (tea waste)	33.81
Atenolol	Granular activated carbon	4.00
Bentazon	Calcined hydrotalcite	123.10

Table 2 shows a list of emerging contaminants removed by using different adsorbent materials having varying adsorption capacities. For instance, polyaniline nanotubes exhibited maximum adsorption capacity (i.e., 0.002 mg.g-1) for the removal of perfluoro octane sulfonate, whereas, activated carbon exhibited the least adsorption capacity (i.e., 1651 mg.g-1) for the removal of 4-chlorophenoxyacetic acid and clofibric acid.

### 3.2 Removal of pharmaceuticals by using membrane technologies

Among various physical treatment methods, membrane treatment stands out because of its remarkable efficiency in eliminating a range of contaminants from water [66]. Commonly microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) membranes are used in water treatment [67]. This method is promising in terms of removal efficiency; however, high cost and energy consumption make it an out of reach option for developing countries. Various researchers studied the removal of pharmaceuticals using UF and MF but the results were not satisfactory [68, 69]. Apart from the membrane type, the nature of pharmaceuticals also dictates the contaminant-removing capacity. For instance, the UF membrane is effective in removing non-steroidal anti-inflammatory drugs (NSAIDs), however, results are underrated for polar phthalate esters [70]. NF membranes exhibit higher removal efficiency as compared to MF and UF due to their small pore size [71]. Taking into consideration different membrane characteristics and working conditions, NF and RO can remove 40% to 100% of emerging contaminants [72].

### 3.3 Removal of pharmaceuticals using Fenton or Fenton-like process

Fenton or Fenton-like processes are the types of advanced oxidation processes (AOPs) that use hydrogen peroxide to produce hydroxyl radical ( $\cdot\text{OH}$ ) for oxidation of the contaminants in an aqueous medium [73]. Previously, this process was applied for the removal of amoxicillin and a remarkable degradation rate was achieved, however, bioactive photoproducts of amoxicillin were formed [74]. Similarly, Sun, et al. [75] reported 100% degradation rates for norfloxacin, cephalexin, and ofloxacin via photolysis with hydrogen peroxide. Table 3 shows the efficiencies obtained by using Fenton/Fenton-like processes for the removal of ECs [76].

Table 3: Studies conducted on the removal of ECs by using Fenton/Fenton-like processes.

Emerging contaminants	Process	Removal efficiency (%)
Levofloxacin	Photo-Fenton	80
Tetracycline	Photo-Fenton	80
Carbamazepine	Electro-Fenton	100
Pymetrozine	Electro-Fenton	90
Dimethyl phthalate	Electro-Fenton	96
Bisphenol A	Photo-Fenton	75

Table 3 shows that reasonable efficiencies have been achieved for the removal of a range of ECs by using these processes. Out of all processes, the Electro-Fenton process has shown promising efficiency i.e., 100%, 90%, and 96% removal of carbamazepine, pymetrozine, and dimethyl phthalate, respectively.

## 4 Practical Implication

The information provided in this review can be utilized in choosing appropriate treatment options under given circumstances. Furthermore, this review also holds its significance as a single-read source for upcoming researchers in the



domain of arsenic and emerging contaminants removal. Comparative analysis of various treatment methods has disclosed the need for integrated treatment technologies to formulate a universal solution for treatment. This review is helpful for upcoming researchers, practitioners and water engineers striving for the effective removal of understudy emerging contaminants.

## 5 Conclusion

This review concludes that health impacts of arsenic contamination have been widely explored, however, a huge gap of obscurity exists when it comes to the health impacts of emerging contaminants. Further, it was found that both natural and anthropogenic sources trigger arsenic contamination; whereas, pharmaceuticals are mostly released into the environment by anthropogenic means. The drastic rise in arsenic and pharmaceutical load in the water has compelled the researchers to investigate various treatment methods. Outcomes disclosed that the selection of suitable treatment technologies depends on user constraints and prevailing conditions.

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