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# Arsenic removal technologies for middle- and low-income countries to achieve the SDG-3 and SDG-6 targets: A review

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# ABSTRACT

Contamination of arsenic (As) in groundwater has increased across the world with prominence in the middle- and low-income countries. The United Nations 'Sustainable Development Goals' (SDG's)- 'good health and wellbeing' (SDG 3) and 'safe and clean water and sanitation for all' (SDG 6), cannot be achieved without monitoring and remediating 'As' pollution in groundwater. Over 230 million people worldwide are affected due to arsenic-contaminated drinking water. More than 200 articles discussing the 'As' contamination, toxicity and cost effective technology were reviewed in this study focusing on economic status of the affected nation as per World bank report. Cost budget analysis suggested that chemical oxidation followed by precipitation (0. 043-0. 076 US  $(m^3)$ ), hybrid treatment technologies (0. 15-0. 17 US $(m^3)$ ) and biological oxidation (0. 2 US $(m^3)$ ) can be applied in low income countries through community based models to mitigate the health problem related to As contamination in order to achieve SDG 3 and SDG 6 targets. This study recommends further research on budget friendly 'As' remediation systems and policy level interventions in the affected nations to cater safe drinking water to all.

# 1. Introduction

Elevated concentrations of As in the groundwater is a serious concern in many parts of the world especially low and middle income countries such as India, Bangladesh, China, Cambodia, Nepal and, Vietnam (Smedley and Kinniburgh, 2002). Over 85% of world's population is dependent on the groundwater for their drinking water source (World Bank, 2012; Shaji et al., 2021). Arsenic is a geogenic pollutant, originating from the minerals in the rocks. The distribution of As is more prevalent in the areas of active plate tectonics, magmatism and associated hydrothermal activity. But 'As' pollution is not limited only to tectonically active areas. One of the examples is the sedimentary alluvial fan of lower Ganges- Brahmaputra- Meghna basin, the home of 400 million people. Here As is a geogenic pollutant found in the semi-confined/confined/ unconfined aquifer located between the Pleistocene terraces including the Barind (North Bengal), the partially weathered gray silts and clay floodplains and Chottanarpur formations in the West.

Over 230 million populations are suffering from As contamination in groundwater (Shaji et al., 2021). Among these, 78% of this exposure is reported in south Asian developing countries (Rajendran et al., 2021). Current literature indicates that the groundwater in India, Bangladesh, China, Cambodia, Nepal, Vietnam, Argentina, Bolivia, Chile, Mexico, and Brazil have been contaminated with As (Ravenscroft et al., 2011; Singh et al., 2015; Sarkar and Paul, 2016; Bindal and Singh, 2019). As per World Bank report, all these major effected nations belong to the category of lower and middle income countries (World Bank, 2020).

UN Sustainability Development Goals addresses the major issues of development and envision a green, sustainable future by 2030. Arsenic pollution and exposure to human population is one of the major hurdles to achieve the targets of 'Good health & wellbeing' (SDG-3) and its major target "reduce illnesses and deaths from hazardous chemicals and pollution (target 3. 9: SDG goals)". It is also vital to manage the As pollution to achieve SDG-6 (clean water and sanitation for all) and its

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major target 6. 1: Safe and affordable drinking water. The dependency on groundwater is difficult to circumvent in middle-low income countries with high population density. Hence, viable As removal techniques are mandatory if policy makers and engineers can even try to achieve the SDG-6 targets by stipulated deadline of 2030.

Sources of As contamination are mainly sulfide minerals (chalcophilics) such as arsenopyrite (FeAsS), realgar (AsS), niccolite (NiAs), cobaltite (CoAsS), hydrothermal water, volcanic ash, and iron oxyhydroxide/oxide as weathering products ((Masuda, 2018); Reimann and Caritat, 2012; 2742 Uddin and Jeong, 2020). Geological processes such as hydrothermal activities, ore depositions and sediment weathering naturally contribute to As contamination in groundwater (Zkeri et al., 2018; Jeelani et al., 2020; Wei et al., 2021).

Commonly used As removal technologies are ion exchange, membrane filtration, reverse osmosis, adsorption and chemical and biological remediation. Most of these technologies are expensive, complex and not sustainable for long term application especially in low and middle income countries (Abejón et al., 2015). Such as chemical oxidation followed by precipitation and adsorption methods requires chemicals and generate As contaminated sludge which disposal adds up extra operational cost, in contrast ion exchange, reverse osmosis, membrane filtration produces less sludge but are expensive due to costly membrane, high electricity consumption and complex method and demands experts for proper maintenance. Biological methods of As removal is not very effective for high As contaminated water, although it is suitable for the water contaminated with low concentration of As. The search of the suitable As removal technology is still a major challenge among researchers, scientists, and policy makers (Amen et al., 2020).

Through this review, we present the sources, occurrence of As contamination in groundwater. We have elucidated how As chemistry affects contamination of groundwater resources. An appraisal of recent methods to remediate As from drinking water that are being used across the globe along with limitations associated with the technologies and challenges encountered are presented in detail. This review also presents a cost effective analysis to decide the best suited As removal technology

for low and middle income countries in view of achieving SDG goal 3 and 6.

#### 2. Toxicity of As

Arsenic enters in humans either through contaminated drinking 'As' laden water, or by consuming 'As' bio-accumulated food crops (Singh et al., 2015). Contamination of As has impacted human lives by getting bio accumulated, bio-transformed and bio magnified across the food chain ((Chakraborti et al., 2016a,b; Kumari et al., 2018). Arsenic causes oxidative stress by damaging redox-sensitive signaling molecules by metal induced signal transduction pathways leading to the activation of a transcription factor governing the expression of the genes involved in several diseases, including cancer (Mandal, 2017; Hasanuzzaman et al., 2018; Necula et al., 2021). Arsenic is responsible for a variety of complications in the human body such as the immune system, cardiovascular, brain, nervous, hepatic, reproductive and renal functions (Naujokas et al., 2013; Chakraborti et al., 2018) (Fig. 1). Chronic exposure of As can cause Parkinson's, melanosis, leucomelanosis, keratosis, gangrene, dermal irritation, and cancer (Panda, 2015; Dorsey, 2018). The elevated As can accumulate in the hair and nails (Gault et al., 2008; Francesconi, 2010; Wongsasuluk et al., 2021).

Arsenic impact the zinc finger DNA repair proteins- poly (ADPribose) polymerase-1 (PARP-1), playing a significant role in increasing chances of cell mutation which can eventually lead down to carcinogenesis (Zhou et al., 2021). Deposits of As in the dermal layers causing depletion of S-adenosylmethionine pool and causes hypermethylation of Arsenite methyltransferase, that can induce carcinogenesis in skin (Das et al., 2021).

Recent researches have highlighted that impact of As reduces the semen production capacity in males due to inhibition of androgen receptor and ERK/AKT/NF-kB signaling pathways crucial for spermatogenesis (Sengupta et al., 2013; Renu et al., 2018). Arsenic, a major Endocrine disrupting agent, impact the Glutathione reductase enzyme action in the Pentose Phosphate Pathways (PPP) and inhibits the cell

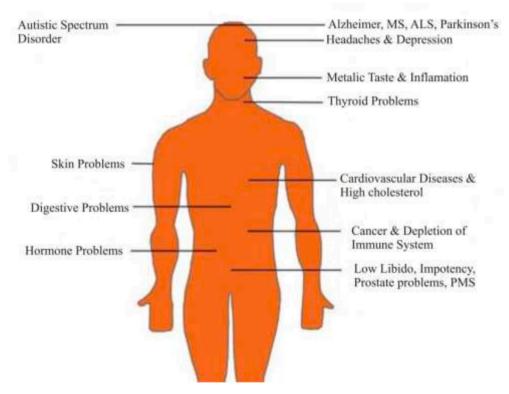


Fig. 1. Long term Arsenic exposure and their associated health impacts on human (modified from Panda 2015)

signaling molecules required for production of endocrine signals (Sabir et al., 2019). Arsenic can be a factor for progression of Parkinson's disease, as it induces imbalances in Ca<sup>2+</sup>pathways during ATP synthesis mechanisms in a neuron due to over production inflammation inducing cytokines such as Interleukin- 1 $\beta$  (IL-1 $\beta$ ), IL-6, Tumor Necrosis Factor -  $\alpha$  (TNF- $\alpha$ ) (Jomova et al., 2010). In cause-effect studies focusing on 'Parkinson's disease' indicates that As have propensity to mutate/ hinder transcription-translation from DJ-1 in the neuron cells by increase oxidative stress in the cell (Raj et al., 2021).

Consuming a high protein diet such as milk, fish, pulses and meat can prevent toxicity to some extent (Mondal et al., 2013). High income countries have resources and technologies to removal of As at source as well as the people can effort sufficient protein diet. Unfortunately marginalized people of low and middle income countries cannot afford the protein rich diet is at highest risk of As toxicity, which again hinder the goal SDG-3. To achieve SDG-3 goals, the removal of As at source through commercial and household level is the primary necessity.

WHO (World Health Organization) has constantly been tracking the As levels in the groundwater. With latest research inputs and increase in the incidence of the toxicity of As and its biomagnification WHO and statutory bodies of different countries have revised the guidelines for the permissible limits (Table. 1). These issues are majorly responsible for inability of nations across the world to effectively manage the problem of As-induced health hazard and without proper remediation technology innovations, it will be impossible to comply with the global SDG-3 targets.

#### 3. Arsenic impacted countries according to economic status

About 90% of As comes from geogenic sources and most of the Asian countries are at high risk of As exposure (Shaji et al., 2021). The toxicity

#### Table 1

Chronology of alterations in acceptable/permissible limits for Arsenic concentration in Drinking water guidelines advocated by different national and international statutory bodies.

Standards	Year (Updated)	Standard limit (μg/l)	Remarks
WHO	1958	200	WHO International Drinking water Standard:
WHO	1963	50	WHO recommend lowering guide value from 200 to 50 µg/l
WHO	1993	10	WHO provisional guideline recommends lowering guide value to from 50 to $10 \ \mu g/l$
European Standard for Drinking water	1998	10	Lowered the acceptable limit. EU. 1998
Standard for drinking water Quality: Ministry of Health of China (GB 5749- 2006)	2006	50	This limit applicable for all drinking water supply sources of urban and rural areas of China
USEPA and National Research council	1974	50	Later the standard proposed to reduce the maximum contamination units from 50 to 10 µg/l in January 2006
US EPA: National Primary Drinking Water Regulations	2009	10	Public health goal is set for zero contamination of As in water
Indian Standard (IS: 10500)	2012	10	Acceptable limit is 10 $\mu$ g/l, while the permissible is upto 50 $\mu$ g/l
Australian Drinking Water Guidelines 6 2011	2017	10	These values are as per National Health and Medical Research Council (NHMRC) of Australian Government

of As through groundwater source is a challenge especially for the low and middle income countries due to limited resources and funds. Arsenic concentrations and sources reported in different countries are listed in the Table 2. Although some of the high income countries are reported upto 10000  $\mu$ l of As, the exposed population are very less in comparison to Asian countries (Rahaman et al., 2021). As exposed population are highest from the Asian countries (> 200 million) followed by America (2.5 million), Europe (1. 2 million) Africa (0. 5 million), and Australia (0.001 million) (Kumar and Ghosh, 2021; Shaji et al., 2021). More than 30 Asian countries are reported high As concentration in groundwater out of which severely As impacted countries are Bangladesh, India, China, and Pakistan (Table 2).

On the basis of mortality rate, the highest As toxicity impacted country is Bangladesh where >40% of groundwater sources are As contaminated and about 21. 4% of all deaths occur because of high concentration of As (> 10  $\mu$ g/l) in drinking water (WHO, 2018). About 1. 47 crore Indian are at high risk of As induced cancer out of which over 1 lakh deaths is also reported due to chronic exposure of As through water (Dutta et al., 2019). Agenda for Sustainable Development on holistic management of drinking water services and monitoring of drinking water quality by 2030, cannot be possible till the global profile of arsenic contamination not monitored and managed (Johnston, 2016). The regular monitoring of As contamination and proper management is necessary to achieve SDG 3 and SDG 6 goal.

#### 4. Arsenic removal technologies

Extensive research on the As removal from water has been carried out across the world (Singh et al., 2021). Most of the studies have been carried out in the laboratories, while a few were performed in the field. However, many of these methods have shown success at pilot scale and do not work in field conditions (Vangronsveld et al., 2009; Baig et al., 2015; Kurz et al., 2020). For example, the membrane filtration method, although effective in As removal, is a costly and complex procedure. Ion exchange is very effective at small scale for As removal, however at large scale it is expensive and has high maintenance cost (Höll, 2010). Adsorption method is also expensive at pilot scale due to high cost of adsorbent, and it is difficult to separate As in presence of other competitive ions in water (Siddiqui and Chaudhry, 2017). Most of the expensive technology is well adopted by the high-income countries around the world; however, for the middle- and low-income countries it is not affordable. Coagulation with metal salts and lime followed by filtration is the widely used method and significantly economic and efficient for As removal from water (Ge et al., 2020). Microbes are used to remove As from water, however very few study highlighted the usage of microbes for As from contaminated water at pilot scale (Laroche et al., 2018).

The methods based on reverse osmosis, ion exchange, adsorption through nanoparticles, membrane filtration, have shown significant efficiency in As removal from water, but are complex, require expertise and are economically not viable. Several Asian low- and middle-income countries are severely affected by geogenic pollution which requires easy and economically feasible options. Various As removal techniques and their advantages and disadvantages are depicted in Table 3. Various properties of this element and its valence states are exploited to devise an effective protocol to decontaminate the water.

#### 4.1. Removal of As by chemical oxidation

In groundwater and in anoxic conditions, As is predominant as arsenite [As (III)] at neutral pH (Smedley and Kinniburgh, 2002; Kanel et al., 2005; Drahota et al., 2009). According to Singh et al. (2015) it exists as oxyanion with a neutral charge, H<sub>3</sub>AsO<sub>3</sub> which has a very low affinity towards mineral surfaces and thus remains in soluble form. In oxidizing environments, As exists in pentavalent state *i. e,* arsenate [As (V)] having negative charge, H<sub>2</sub>AsO<sub>4</sub> or HAsO<sub>4</sub><sup>2-</sup> (Kang et al., 2000).

# Table 2

Arsenic concentration (µg/l) in groundwater reported in the different countries according to their economy list (World Bank 2020).

Economy	Countries	Location	Arsenic Concentration	Sources	Refs.
High income of America of America Chile New Zealand Mexico Finland Canada Germany France		Western USA (Alaska, Arizona, California, Hawaii, Idaho, Maine, Nevada, Oregon, and Washington)	1-48000 24,300	Older basin-fill sediments due to dissolution of sulphide and pyrite minerals	Welch et al., 1988 Shaji et al., 2021, Flanagan et al., 2015
	Chile	Northern and central parts of Chile	800-27000	Aquifer contaminated due to quaternary volcanogenic sediments	Dauphiné et al., 2011, Corradini, et al. 2018
		Waikato region Lagunera, Valle del Guadiana,	8. 0-9080. 0 0-2400	Geothermal waters Volcanic sediments	Lord et al., 2012 Shaji et al. 2021
	Finland	valle da Zimapan South-west Finland	0-2230	Weathering of minerals basically Arsinopyrite and As- sulphide	Kurttio et al. 1998; Pedretti et al. 2019
	Western Quebec Eastern and Southern Germony	0. 2-326. 4 0-550	Bedrock weathering As contamination through alluvial sandstone and sediments affects the shallow aquifer	Bondu et al. 2017 Heinrichs and Udluft, 1999; Shaji et al. 2021	
	Alpine/Mediterranean Var River watershed and partially remediated mining site in Southern France	0-263	As laced bedrock	Shaji et al., 2021, Barats et al, 2014	
	Romania	Transylvania	1-176	As mineral rock weathering	Gurzau and Gurzau, 2001
	Japan	Fukuoka Southern Japan (Kyushu island)	0. 001-0. 293 25,700	Volcanic sediment, Holocene coastal sands	Kondo et al., 1999 Shaji et al., 2021; Even et al., 2017
Upper middle income countries	Brazil	Ribera Valley, Amapa State, Rio das Valihas, Minas, Gerais, Rondonia State, Amazon	100,000	Aquifer gets contaminated through As by sulphide and gold ore	Matschullat et al., 2000; Shaji et al., 2021
countries Argentina Thailand China Iran	Argentina	Eastern, North eastern North central Argentina	14,969	Tertiary quaternary volcanic deposits	Robles et al., 2016; Shaji et al. 2021
	Thailand	Chaco-Pampean plain, Pampas, Cordoba Nakhon Si Thammarat Province,	10–5300 100-3810 1. 25-5114	Shallow (alluvial) aquifer contaminated due to	Nicolli et al., 2012 Nicolli et al., 1989 Williams et al., 1996
		Ronpibool,	1-5000	mining process Water contaminated by tin mining waste	Choprapwon and Porapakkhar 2001
	China	Inner Mongolia, Xinjiang, PR China Shanxi, PR	1-2400 0. 05-850 0. 03-1. 41	As-Fe rock weathering, Holocene alluvial sediment	Guo et al. 2001 Yinlong, 2001 Yinlong, 2001
	Iran	North-west, North-east, South- east, and South-west of Iran, Rafsanjan plain	4-1061	Volcanic mountains and copper mines, weathering of sediments	Hamidian et al., 2019; Rahnamarad et al., 2020
	Mexico	Lagunera region	8-624	Volcanic sediments	De Razo et al., 1990
	Russia	Republic of Dagestan,	1-500	Geothermal waters	Abdulmutalimova et al., 2019
Peru	Karelia, Locumba River; Western Amazonia	2–89. 3 500-700	Water of the flooded mine "Arsenic" Unconfined aquifers composed of pure sand	Cherkasova et al., 2021 Sancha and Castro, 2001	
	Indonesia	West Java	10-350	River and Aquifer	Fauzi et al., 2017
	South Africa	Northern Cape Province	<20-252. 86	Sulphide minerals, goldmines	Abiye and Bhattacharya, 2019
	Brazil	Minas Gerais State	0. 5- 40. 1	Water contaminated by gold mining	Bidone et al., 2016
Lower middle income	India	Districts of East India including (West Bengal), Assam (Barak Valley)	3-3880	Arsenic-rich sediments; Rock-water interaction; via arsenic-contaminated alluvial aquifers; partially weathered gray silts and clay floodplains and	Mandal et al., 1996; Shaji et a 2021; Thambidurai et al, 2013 Mahanta et al, 2015
countries Ghana Vietnam		Bihar, Jharkhand, Uttar Pradesh, (Upper and middle Ganga Plain)	10-3192	chottanarpur structure; aquifer semi-confined between the Pleistocene terraces	Saha et al. , 2010; Alam et al. 2016; Hundal et al., 2007; Wu
		Punjab, (Northwest India) North-eastern India including Bishnupur district, Manipur	50-986		et al. , 2021 Oinam et al. , 2011; Saurav et a 2015
		Southern and central Ghana	0-4500	Aquifer in confluence with As bearing rocks near gold mines	Buamah et al., 2008; Shaji et a 2021
		Mekong delta Hanoi	0-1470 1-3050	Deep aquifer Arsenic-rich sediments	Erban et al., 2013 Berg et al., 2001
	Cambodia Nepal	Mekong River floodplain Nawalparasi district	1-1340 10-1338	Sediment aquifers	Buschmann et al., 2007 Yadav et al., 2014
	Nepal Bangladesh	Nawaiparasi district Chapai Nawabganj district of	10-1338 2. 34 -586. 96	Aquifer contaminated with As due to oxidation- reduction process of iron oxides and pyrite Holocene alluvial sediments, organic matter,	Islam et al., 2019
	Dellist	Bangladesh	0 4 100	reducing, salinity, As rich minerals	
	Pakistan	Vehari	0. 4- 132 1. 0-105. 3	Arid and semiarid region contaminated with As; Quaternary sediments	Shahid et al, 2018 Shah et al., 2020
	Sri Lanka	Mannar island	0. 60-34. 0	Reductive dissolution of Fe-Mn oxides and oxy- hydroxides coated on sandy aquifer materials	Bandara et al., 2018

Oxidation leads to conversion of soluble Arsenite to partially soluble Arsenate and subsequently precipitation of Arsenate (Jain and Ali, 2000; Bolan et al., 2014). The redox system of As (III) and As(V) and can be stated as follows:

 $H_3AsO_4(aq) + 2H^+ + 2e^- \rightarrow H_3AsO_3(aq) + H_2O(l)E_0 = +0.56V$  (1)

The standard oxidizing potential for As (III) to As(V) is less than the

Water treatment Techniques	Various processes used	Merits	Demarits	References
Chemical oxidation and precipitation	Oxidation through manganese dioxide, chlorine, permanganate, ozone, and ferrate	Chemical are easily available, low cost with simple operation	Slow process, sedimentation and filtration is required, produce high volume of liquid waste which increase the operational cost	Zakhar et al., 2018
Coagulation and flocculation	Aluminum, Iron, titanium chloride, zirconium chloride based coagulants	Materials easily available, process is not complex, not required skilled technician	Produce large volume of As bearing sludge, which required proper disposal and this adds up cost	Höll, 2010
Electrocoagulation	Iron electrode	100% removal is possible at laboratory scale	Sludge management needed	Kabir and Chowdhury, 2017
Biological treatment	Biological oxidation of As (III), arsenic removal through microbes, microalgae and fungi	Generated sludge can be used for waste water treatments, Cost effective	Treated water needs tertiary treatment before supply	Ghosh et al., 2019
Adsorption	Aluminum based sorbents, activated carbon, iron based sorbents, clays and soils,, zeolites and other miscellaneous adsorbents	Comparatively high removal efficiency	Suspended salts affects arsenic removal efficiency; The adsorption efficiency highly dependent on the water quality	Luong et al., 2018
Membrane filtration	Membrane distillation, microfiltration, nanofiltration, ultrafiltration	Best available technology for water treatment to remove arsenic, High removal efficiency	Wastage of huge amount of water with toxic waste water	Dutta et al., 2012
Nanoparticles	Iron, titanium and zirconium based nanoparticles	Simple handling, less working area required for treatment processes	For better efficiency it is required to connect photochemical process	Siddiqui et al., 2019
Ion exchange	Using different ion exchange membranes	High efficiency and commercially available	Not suitable for high TDS water. Pretretatment required. Costly and regeneration of membrane is difficult	Mohanty, 2017

oxidizing potential of Ferric [Fe(II)] to Ferrous [Fe(III)]. However, the oxidation rate of arsenite is relatively much less than Fe (II) and hence arsenite can sometimes exist in a meta-stable state even in oxidizing environments (Ahmad et al., 2018).

The popular oxidizing materials for oxidizing arsenite to arsenate are chlorine, chlorine dioxide, chloramine, ferrate, hydrogen peroxide, manganese oxide, ozone, and permanganate (Kim and Nriagu, 2000; Manning et al., 2002a, b; Lee et al., 2003; Ghurye et al., 2004 Dodd et al., 2006; Sorlini and Gialdini, 2010). Fig 2 shows the role of KMnO<sub>4</sub> and FeCl<sub>3</sub> in oxidation of arsenite by the advanced oxidation-coprecipitation-filtration (AOCF) method.

#### 4.1.1. Chemical oxidation using manganese dioxide

Manganese dioxide is a strong oxidant in an environment which can reduce the mobility of As, Fe, Co, Cr, and organic matter (Oscarson et al., 1980; Crowther et al., 1983; Eary and Rai, 1987; Li et al., 2010). Feng et al (2006) synthesized three Mn oxide minerals namely, birnessite, cryptomelane, and hausmannite which generally occur in soil and sediments. Depending on their respective crystal structure, composition and surface properties, they have different abilities for As (III) oxidation. Cryptomelane has a tunnel structure, while birnessite has layered structure, and hausmannite shows a lower oxide structure. The highest ability of As(III) oxidation has been exhibited by cryptomelane followed by birnessite and hausmannite respectively (Feng et al., 2006).

The oxidative reaction of As (III) with cryptomelane (or birnessite) is as follows:

(1). At lower pH condition:

$$2(MnO_2)_x(s)+2H_3AsO_3(aq.)+H^+ \rightarrow H_2AsO_4^-(aq.)+HAsO_4^{2-}(aq.)+2Mn^{2+}(aq.)+2(MnO_2)_{x-1}(s)+2H_2O(l)$$
(2)

#### (2). At higher pH condition:

$$\label{eq:2} \begin{split} &2(MnO_2)_x(s) + 2H_3AsO_3(aq.) \rightarrow H_2AsO_4^-(aq.) + HAsO_4^{2-}(aq.) + 3H^+(aq.) + 2 \\ &(MnO_2)_{x-1}.MnO(s) \end{split}$$

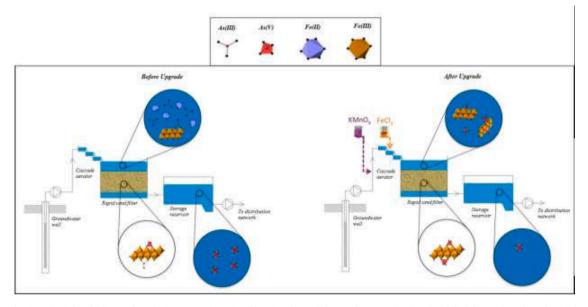


Fig. 2. Principle of advanced oxidation-coprecipitation-filtration (AOCF) for As decontamination (modified from Ahmad et al. 2018).

In the above equations  $(MnO_2)_x$  is denoted for Mn oxides *i. e*, cryptomelane and birnessite. With an initial increase in pH value, the amount of As(III) oxidized was first decreasing. But as the pH was continuously increasing, the oxidation amount of As(III) by Mn oxide has increased remarkably.

Manning et al. (2002a) experimented As removal through birnessite. For this process a conventional stirring technique and an extending X-ray adsorption fine structure spectroscopy (EXAFS) both have been used. The stirred reaction experiment showed, the oxidation of As(III) was taking place before the adsorption of Arsenate on MnO<sub>2</sub> solid phase. EXAFS study confirmed As(V)- MnO<sub>2</sub> complex as a 'bridged complex' formed at MnO<sub>2</sub> crystal corners as well as on the interlayer parts (Mohan and Pittman Jr, 2007; Su and Puls, 2008).

#### 4.1.2. Chemical oxidation using chlorine-based oxidants

Free available chlorine (FAC) can oxidize As (III) under all the tested conditions (Sorlini and Gialdini, 2010). The oxidation reaction between As (III) and FAC is as follows:

$$H_{3}AsO_{3}(aq.) + NaClO(aq) \rightarrow H_{2}AsO_{4}^{-}(aq) + Na^{+}(aq) + Cl^{-}(aq) + H^{+}(aq.)$$
(4)

In the above reaction, oxidation of 1 mole of As (III) requires 1 mole of NaClO (hypochlorite), which is equivalent to 2 moles of Cl<sub>2</sub> (Clifford and Ghurye, 2001). Experiments performed by Dodd et al. (2006) described that only 0. 1 mg/l of Cl<sub>2</sub> dose as FAC is adequate for the depletion of As (III) in a real water samples spiking 50  $\mu$ g/l As(III) level to <1  $\mu$ g/l of As(III) within 10 s.

Chlorine dioxide ( $ClO_2$ ) was also used for the oxidation of As (III), but it was not very effective (Clifford and Ghurye, 2001). This study suggested that only 20 to 30% of oxidation took place in the 3-fold stoichiometric dose of  $ClO_2$ , and after that oxidation nearly stopped. However, with an increase in pH, the oxidation reaches from 30% to 50% (Sorlini and Gialdini, 2010). It appeared that the initial Arsenite concentration does not increase oxidation efficiency significantly, as the researchers got similar results at 300 mg/l of As (III). The reaction for oxidizing As (III) using  $ClO_2$ :

$$H_3AsO_3(aq.) + 2ClO_2(g) + H_2O(l) \rightarrow H_2AsO_4^-(aq.) + 2ClO_2^-(aq.) + 3H^+$$
 (5)

Ghurye and Clifford (2004)) attempted an experiment using a preformed NH<sub>2</sub>Cl. It was observed that there was no As (III) oxidation taking place. But the *in-situ* formed NH<sub>2</sub>Cl oxidized As (III) to about 40%.

$$H_3AsO_3(aq.) + NH_2Cl(s) + H_2O(l) \rightarrow HAsO_4^-(aq.) + NH_4^+(aq.) + Cl^-(aq.) + 2H^+$$
(6)

Iron with concentration of 0. 3-3 mg/l, and manganese with 0. 2 mg/l, had shown very little effect on As(III) oxidation process (Bundschuh et al., 2021). But sulfides (1-2 mg/l) slowed down the process to a greater extent. In presence of sulfide, the oxidant dose used was 3-10 times the stoichiometric requirement to oxidize As(III) (Clifford and Ghurye, 2001)

# 4.1.3. Chemical oxidation using potassium permanganate

Sorlini and Gialdini (2010) carried out an experiment on the As oxidation through potassium permanganate. Their research suggested Potassium Permanganate can oxidize As(III) up to 95 to 98% at the initial 50 mg/l of Arsenite. However, the increased doses of KMnO<sub>4</sub> enhanced the oxidation efficiency of Arsenite up to 100%. This oxidant worked well at the initial 300 mg/l of As(III) concentration, but this required increasing the oxidant dose to get higher efficiency. However, there was no change in oxidation yield when the pH changed. The reaction is as follows:

$$H_3AsO_3(aq.) + 2MnO_4^-(aq.) \rightarrow 3H_2AsO_4^-(aq.) + 2MnO_2(s) + H_2O(l) + H^+$$
 (7)

#### 4.1.4. Chemical oxidation using ozone

Ozone acts as a rapid oxidant. It took only 15 s to oxidize > 95% of arsenite. However, the oxidation rate suddenly reduced due to the presence of total organic matter and sulfate (Ghurye et al., 2004a; Khuntia et al., 2014). It is also suggested that the higher dose of  $O_3$  is needed to oxidize As(III) in groundwater which is already enriched in total organic carbon. The following reaction takes place between  $O_3$  and As(III):

$$H_3AsO_3(aq.) + O_3(g) \rightarrow H_2AsO_4^-(aq.) + O_2(g) + H^+(pH=6.5)$$
 (8)

 $H_3AsO_3(aq.)+O_3(g) \rightarrow HAsO_4^{2-}(aq.)+O_2(g)+2H^+(pH=8.5)$  (9)

With the help of pure oxygen ( $O_2$ ) and air as oxidants, about 57% and 54% of As(III) was oxidized respectively in the near neutral pH range (Kim and Nriagu, 2000).

#### 4.1.5. Chemical oxidation using ferrate

Potassium ferrate [Fe(VI)] is highly stable, environment friendly, and has a very high oxidizing power. The bi-product of the oxidant is Fe (III), which is non-toxic in nature (Sharma, 2002). The oxidation of arsenite using ferrate with a stoichiometric ratio of 3:2 [As(III):Fe(VI)] has also proven as an efficient method (Wang et al., 2021). Researchers also stated that the concentration of 'As' was lowered to  $<50 \ \mu g/l$  from initial concentration of 517  $\mu g/l$ 'As', using just 2 mg/l of Fe(VI) (Lee et al., 2003). However, in order to get better results the pH of the water should be 4 and time for optimum reaction is 5 min. The reaction is given below (after Singh et al. 2015).

# $3As(OH)_3(s) + 2FeO_4^{2-}(aq.) + H_2O(1) \rightarrow 3AsO_4^{3-}(aq.) + 2Fe(OH)_3(s) + 5H^+$ (10)

Oxidation with potassium permanganate, ozone and ferrate has shown high efficiency for As removal in comparison to oxidation with chlorinated compounds. The chemical oxidation and precipitation is the most convenient, feasible and effective method. However, the biggest disadvantage is the disposal of As contaminated low dense sludge volume. The disposal of liquid waste makes this process costly (Kabir and Chowdhury, 2017). This process needs chemicals and required quite trained operators. Precision is most important to achieve maximum removal efficiency.

#### 4.2. Coagulation and flocculation

Coagulation and flocculation basically depend upon the coprecipitation of both inorganic oxides of other metals and the insoluble arsenates (Mohindru et al., 2017). The process involves the addition of metal-based coagulants such as ferric chloride, ferric sulfate and alum to the contaminated water (Sancha, 2006; Rathi and Kumar, 2021). For maximum As removal efficiency, the coagulant is mixed homogeneously throughout the aqueous medium. The metal-based coagulant e. g. FeCl<sub>3</sub> hydrolyzes itself in water and forms positively-charged ferric hydroxide Fe(OH)<sub>3</sub> (Wang et al., 2021). As(III) is pretreated with some oxidant to convert into As(V), it is a necessary step in the elimination of As from contaminated water. As(V) is an anion and it is attracted to the positively charged Fe(OH)<sub>3</sub> flocs (Fig. 3). In the whole process of coagulation and flocculation, pH, As speciation, coagulant dose, and the agitation speed are the major parameters (Sancha, 2006).

Aluminum sulfate (alum) is the most widely used coagulant for water treatment in United States of America (Hering and Elimelech, 1997) and in the developing nations. Addition of coagulants increases the As(V) removal. The coagulants accelerate the elimination of Arsenate from the aqueous solution by transforming it into insoluble products (Wickramasinghe et al., 2004. The more 'alum' dose is used, the more aluminum oxide is formed, which provides a greater surface area for the adsorption of As(V) (Baskan and Pala, 2010). This work suggested 100% removal of arsenate at initial As concentration of 500 and 1000  $\mu$ g/l, at the

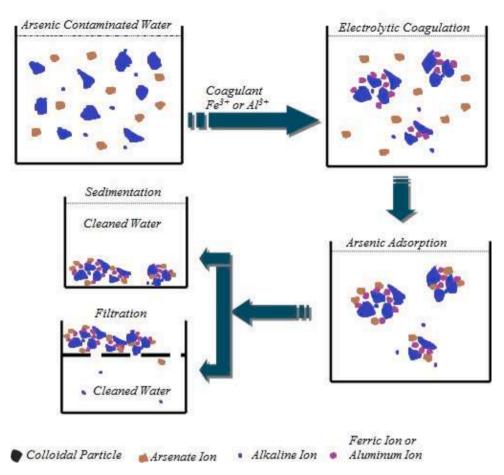


Fig. 3. Arsenic removal mechanism through coagulation and flocculation technique (modified from Song and Gallegos-Garcia 2014.

coagulant doses of 42 to 56 mg/l, respectively. However Matsui et al. (2017) reported 30 mg/l of alum can show the maximum efficiency to precipitate arsenate at a pH value of 7-8 (Habuda-Stanić and Nujić, 2015).

Another coagulant iron salt (FeCl<sub>3</sub>) also studied and found effective in order to remove As from contaminated water (Ghosh et al, 2003). It was detected that both arsenite and arsenate were eliminated from aqueous media with a FeCl<sub>3</sub> of 40 mg/l within a pH range of 6–7. 5. But with the increasing pH value, As(V) removal efficiency reduced very sharply. However, the presence of anions like Cl<sup>-</sup> and NO<sub>3</sub><sup>-</sup> did not significantly affect the coagulation process but the removal efficiency was remarkably low due to SO<sub>4</sub><sup>-</sup>ions. The co-precipitation reaction for different inorganic As species are as follows (Ghosh et al, 2003):

I. As(V) separation-

$$\equiv \text{Fe-OH}(s) + H_2 \text{AsO}_4(aq.) \rightarrow \text{Fe-H}_2 \text{AsO}_4(aq.) + H_2 O(l)$$
(14)

II. As(III) separation-

$$\equiv \text{Fe-OH}(s) + H_3 \text{AsO}_3(aq.) \rightarrow \text{Fe-H}_2 \text{AsO}_3(aq) + H_2 O(l)$$
(15)

where  $\Box$ Fe-OH represents nucleation of precipitation on the solid surface.

Based on the surface complexion model, adsorption and coagulation were compared as it is evident that adsorption is an important mechanism that governs the removal of As during coagulation (Ghosh et al., 2003; Jain and Singh, 2012). However, the removal efficiency of FeCl<sub>3</sub> decreased in the presence of total organic carbon at pH 8-9 (Henke, 2009). Direct filtration technique after the coagulation achieved more than 95% removal efficiency at pH 7 and FeCl<sub>3</sub> dosage of 2 mg/l. Groundwater samples from US and Bangladesh were evaluated for coagulation and filtration by using FeCl<sub>3</sub> and (Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>)

(Wickramasinghe et al., 2004).

Sancha (2000) investigated the As removal efficiency of Titanium chloride (TiCl<sub>3</sub>) as a coagulant. It was reported that both As(III) and As (V) removal were greatly dependent on pH i. e, with the increasing pH As(V) removal efficiency lowered. However, the maximum removal efficiencies of As(V) and As(III) were not more than 49% and 42%, respectively at optimum dosage and pH conditions (Mohindru et al., 2017).

Coagulation and flocculation process ideal for the water rich in Fe and Mg. This process needs very nominal capital investment (Baigorria et al., 2021). This method can be installed at larger scale –community level to supply As free water. Although it remove As upto 95% from contaminated water, alike chemical oxidation and precipitation this process produces As contaminated sludge and disposal of toxic sludge increase the running cost.

#### 4.3. Electrocoagulation

Electrocoagulation is an alternative to the conventional coagulation and filtration techniques. Electrochemical reactions that occur in cathode and anode also include *in-situ* generation of coagulants or flocs (Santos et al., 2015; Ali et al., 2013; Weerasundara et al., 2021). When electrocoagulation was compared to chemical coagulation using FeCl<sub>3</sub>, it was clear that electrocoagulation had better As removal efficiency (Lakshmanan et al., 2010). This technique could overcome the drawbacks of the conventional As removal methods such as low As removal efficiency, time consuming, high operating cost and toxic sludge production (Khandegar and Saroha, 2013; Sahu et al., 2014; Song et al., 2017; Demirbas et al., 2019; Alka et al., 2021).

Metal electrodes mainly of iron or aluminum which are involved in

electrochemical reactions and metallic cations like Fe(II), Fe(III), or Al (III) are generated due to the oxidation reaction on anode. Initially, monomeric Fe or Al ions form FeOH<sup>+</sup>, FeOH<sup>2+</sup>, Fe(OH)<sup>+</sup><sub>2</sub>, AlOH<sup>2+</sup>, Al  $(OH)_2^+$  and polymeric Fe or Al ions form  $Fe(H_2O)_5OH^{2+},\ Fe$  $(H_2O)_4(OH)_2^+$ , Fe $(H_2O)_8(OH)_2^{4+}$ , Al<sub>6</sub> $(OH)_{15}^{4+}$ , Al<sub>7</sub> $(OH)_{17}^{4+}$ , Al<sup>13</sup> $(OH)_{34}^{5+}$ . These ions subsequently form a series of iron or aluminum hydroxides/ oxyhydroxides such as Fe(OH)<sub>2</sub>, Fe(OH)<sub>3</sub>, Al(OH)<sub>3</sub>, with large surface area to adsorb the dissolved contaminants (Kobya et al., 2003). pH acts as an important factor influencing the achievement of the electrocoagulation process. However, the pH depends on the type of electrode used and also on the initial pH. Aluminum electrodes with initial pH < 8give higher final pH in contrast initial pH >8 pH reduces the final pH. In iron electrodes the final pH is always higher than the initial pH. Kobya et al. (2011) performed an experiment to find out the pH at which maximum As removal efficiency can be achieved in electrocoagulation techniques. The optimal As removal efficiency 99% with iron electrode was achieved at a pH of 6. 5 and with aluminium electrode (37%), the pH was 7 (Kumar et al., 2004).

Presence of phosphate as a competing ion inhibits the removal of As during electrocoagulation process (Wan et al., 2011; Song et al., 2014). It was also reported that the presence of phosphorus slows down Fe (II) oxidation to Fe(III) which leads to the reduction in the formation of complexes. When the effect of silica on As removal was evaluated by Wan et al. (2011), it was suggested that there is no significant effect of dissolved silica on As removal efficiency up to 20 mg/l in electrocoagulation process. But Vasudevan et al. (2010) had previously suggested that silica concentrations from 0 to 15 mg/l in the aqueous solution reduced As(V) removal efficiency.

Electrocoagulation process provides 99% As removal efficiency depending on the electrode used and other parameters such as initial As concentration, pH and presence of other ions in water. The disposal of the sludge generated by the process is one of the disadvantages of this technology. Studies of Babu et al. (2021), suggested the management of As-Fe laded sludge generated by electrocoagulation process. This process needs continuous electricity which also adds up additional running cost, and thus not very effective method to install in rural areas of most low income countries.

#### 4.4. As removal through low cost adsorbents

Adsorption technology has been used worldwide due to its high removal efficiency, sludge free and easy operation and handling. Hence, this technique is widely used for the treatment of different pollutants from the contaminated water (Naz and Chowdhury, 2021). Adsorption process is affected by various parameters such as temperature, pH, nature of adsorbate and adsorbent, contact time, concentration of pollutants and, presence of other competitive ions (Chiban et al., 2016). Variety of adsorbent material have been studied on the As removal from drinking water such as aluminium based sorbents, iron oxides, activated carbon, clays and soils, zeolites, flyash, bismuth impregnated biochar, portland cement biochar and chiston crosslinked with transition metals (Aredes et al., 2013; Chutia et al., 2009; Chowdhury et al., 2021; Anjum et al., 2011; Zhu et al., 2016; Han et al., 2019; Kabir and Chowdhury, 2017; Alkurdi et al., 2019; Pincus et al., 2021). Nowadays, plant based adsorbents become more popular due to their cost effectiveness. Study suggested that about 70% removal of As(III) arsenic was achieved at pH 2. 0 by chars (obtained by rapid pyrolysis of oak bark as a byproduct of bio-oil production) (Mohan et al., 2007). However, the cost effectiveness of the adsorption technology is highly dependent on the cost of adsorbent material. As per literature, low cost adsorbent media, such as 5.7 US\$/kg for activated alumina, 6. 05 US\$/kg for granular ferric hydroxide, and 11. 55 US\$/kg for bayoxid, are significantly effective in the removal of As from water (Callegari et al., 2018). Thus the cost of adsorption can be reduced by selecting adsorbent and regeneration materials.

from contaminated water. It provides > 95% As removal efficiency (Baigorria et al., 2021). The installation is simple at reasonable operation cost. This process also does not required trained operators. Less chemical used in this process, except sometimes pH needs to be maintaining for achieving high removal efficiencies. The ions present in the water can compete with As and thus process become complex. The regeneration is difficult and bacteriological contamination may occur during the adsorption process.

#### 4.5. Ion exchange

Ion exchange is a physicochemical process in which the ions of the solid phase are exchanged with the equal number of ions from the contaminated water (Sarkar and Paul, 2016). In this technique, synthetic resins are used as solid phase for absorbing the arsenic ions very efficiently. Balaji and Matsunaga (2002) evaluated the adsorption performance of As(III) and As(V) using TiO<sub>2</sub>-loaded Amberlite XAD-7 resin. It was prepared by impregnating Ti(OC<sub>2</sub>H<sub>4</sub>)<sub>4</sub> followed by hydrolysis using ammonium hydroxide. Results showed strong adsorption of As(V) from a pH range of 1-5 and of As(III) from pH 5-10. Adsorption capacities for As(V) was 0. 063 mmol/g at pH 4 and for As(III) it was 0. 13 mmol/g at pH 7. Cations such as Na (I), Cd(II), Pb(II) did not show significant effect on arsenic adsorption. Anion exchanger (AE) was prepared using coconut coir pith (CP) for removal of As(V) from aqueous solutions (Anirudhan and Unnithan, 2007). A maximum of 99. 2% As(V) removal was achieved at pH 7. 0 for an initial As(V) concentration of 1 ppm with 2 g/l adsorbent dose.

Ion exchange is one of the promising As removal technology. With advancement of science the process become high in demand in high income countries. This process provides > 99% of removal of As from the contaminated water. Most of the time this process does not require pH adjustment except very few cases such as while using activated alumina pH adjustment necessary to achieve highest efficiency (Clifford and Ghurye, 2002). The process is frequent although complex and needs expensive ion exchange resins which may be main reason why it is not popular among low income countries. Regeneration creates a sludge disposal problem, and frequent regeneration of exhausted resins is required. This technique applicable only for low 'total dissolved solids' thus pretreatment require which adds up extra cost (Zakhar et al., 2018). This process requires careful continuous monitoring, thus required trained workers to operate the treatment plant. The installation cost is also very high if it is installed at community level basis.

#### 4.6. Removal of As through biological methods

Nowadays biological remediation techniques are being used more often in developing countries for arsenic removal (Yamauchi and Takata, 2019; Naz et al., 2013, 2021). The role of microorganisms in removing As concentration has been investigated for decades (Banerjee et al., 2013). Biological remediation includes phytoremediation and bio-adsorption using plants, microbes respectively (Stefanescu et al., 2017; Ilieva et al., 2020). Nigam et al. (2013) investigated the biosorption ability of *Hydrilla verticillata* for uptake of As from water under various parameters like pH, temperature, contact time, sorbent dose and initial As concentration. The results showed maximum As adsorption of 96. 35% with the adsorbent dosage of 0. 5g per 100 ml of water at initial As concentration of 100  $\mu$ g/l for contact time of an hour and at a pH 6 (Nigam et al., 2013; Mehdi et al., 2021). Hassan et al. (2009) used both biological and physicochemical treatment methods for As removal.

Arsenic bioremediation depends on the microbial activity to remove, demobilize or adsorbing As from aqueous solutions different biochemical process such as oxidation, biomethylation, complexation and coprecipitation process (Wang and Zhao, 2009; Bahar et al., 2013). Fig 4 depicts activities of microorganisms causing vaporization, precipitation, dissolution, deposition, and redox reactions.

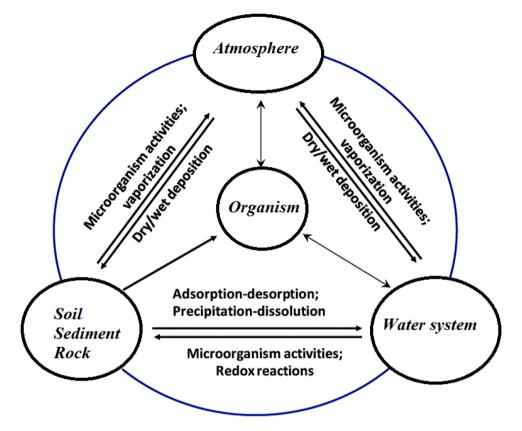


Fig. 4. Mechanism of bioremediation of Arsenic from aqueous solution through micro-organism (modified from Hao et al. 2018).

# 4.6.1. As(III) oxidation through bacteria

Most of the As oxidizing bacteria are isolated from the extreme environments which have high amounts of As (Bachate et al., 2012). This shows that there is a wide distribution of As(III)-oxidizers in the environment (Bahar et al., 2012). The most efficient heterotroph As(III) oxidizer which could oxidize 500  $\mu$ M of As(III) within a contact time of 12 h with initial cell density of 1. 5 × 10<sup>7</sup> cell/mL at pH 5-7 was *Stenotrophomonus* sp. strain MM-7 (Bahar et al., 2012). On the other hand, a chemolithotrophic As(III) oxidizing bacteria *Agrobacterium/Rhizobium* designated as NT-26 was isolated from a gold mine from Australia (Santini et al., 2000; Ito et al., 2012). NT-26 can grow in a minimal salt medium containing As(III) as an electron donor with doubling growth time as 7. 6 h. Species like *Sulfolobus acidocaldarius, Theiomonas arsenivorans* are able to oxidize As(III) at low pH i. e, ( $\leq$  6) (Dastidar and Wang, 2009). But the optimum oxidation of As(III) took place in pH range of 5–7 (Battaglia-Brunet et al., 2011; Mehdi et al., 2021).

Leiva et al. (2014) also suggested the higher efficiency of oxidation by biogeochemical processes in their findings. This method has a high potential of replacing conventional treatment methods of As contaminated water in Asian countries. In a recent study conducted in Italy by Zecchin et al. (2021) it was found out that aquifers contaminated with As had lots of bacterial communities that survived in the polluted waters and these communities have enormous potential to remediate the polluted water. However, studies are at the preliminary stage. Dabrowska et al. (2021) used mushroom waste served as the substrate for the microbial consortia that played a role in remediation of As. Microbial fuel cell found very effective in As removal, it is also effective for removal of organic matter and heavy metals (Tabassum et al. 2021). Since chemicals are not added to the groundwater, it is quite economical and environmentally sound (Katsoyiannis and Zouboulis, 2004a, 2004b; Katsoyiannis et al., 2008). Novel and innovative protocols exploiting the further potential of microbes and other biological organisms using biotechnological and nanotechnology approaches are being developed in various research labs.

#### 4.6.2. Removal of As by microalgae and fungi

Algae display a two-step process in water purification. First step involves the adsorption of metal ions on the cell surface and the second, metal ions perforate into the cells through the cell wall (Bahar et al., 2013; Mehdi et al., 2021). Whereas Fungi help in decontaminating the water by: (i) biosorption of As on fungal surface, (ii) accumulation of As in intracellular spaces and (iii) bio volatilization through some metabolites. Approximately 70% removal of As(III) took place by a common green algae S. abundans when the initial As(III) concentration was 5 mg/l (Jahan et al., 2006). However the maximum uptake of As was observed by Microcvstis aeruginosa when exposed to As(III) and As(V) within 15 days of exposure (Wang et al., 2013). Study suggested that, a wild-type strain Aspergillus niger (fungi) was much more tolerant to As (V) than other fungal strains (Mukherjee et al., 2010). At 25 mg/l As(V) concentration, growth of A. niger increased but they could survive only up to 100 mg/l of As(V) as per Bahar et al., 2013. Detoxification of As through volatilization by methylation has also been studied (Irshad et al., 2021; Tripathi et al. 2020; Verma et al., 2019). However, no study has reported the exact mechanisms behind As detoxification with help of fungi (Irshad et al., 2021).

The biological treatment systems provides significant As removal. The process is less complex and required very less maintenance, does not require much trained operators and suitable for rural areas (Hayat et al., 2017; Zhai et al., 2020). The major disadvantage is it is a slow process of As decontamination from water and suitable for only low As contaminated water. The drinking water treated through microbes, microalgae and fungi needs tertiary treatment before supply. The As removal efficiency is not predictable as this process is highly dependent on the environmental variables such as temperature, and humidity (Lièvremont et al., 2009).

### 5. Discussion

Although many techniques have been developed in the last few years

that show promising results, the sustainability and cost effectiveness of methods is an important issue (Alkurdi et al., 2019). The laboratory protocols show various limitations when applied on the field. The same method applied in different laboratory gives different results, as As removal is highly dependent on the operation, scale of the treatment plants, various parameters, available ions in the water and initial As concentrations. Thus it is difficult to decontaminate the water using similar techniques as the physiographic, climatic, and environmental conditions vary with time and space.

Adopting the hybrid approach such as microbial fuel cell fixing followed by oxidation of As through Fe and Mn oxide can lead to the As removal techniques through an efficient, effective, economic. Research methods using hybrid approaches are now proving popular for the decontaminating As in drinking water. Studies of Maity et al. (2021) also suggested that the hybrid (chemical/adsorption and biological treatment) can be the best As removal techniques in context of cost effectiveness, efficiency and sustainable As remediation strategy. Study of Katsoyiannis et al., 2014 also suggested that hybrid methods such as preoxidation with biological method followed by GFH/adsorption and ozone oxidation followed by chemical precipitation are cost effective and more feasible in comparison to most of the methods (Table 4).

Process complexity, hazardous materials handling, and waste disposal considerations can also have a major influence on the choice of a process. The application of low cost As treatment process can be an effective tool to save millions from As toxicity in middle and low income countries. The cost benefit analysis suggested that chemical coagulation (0. 043 US\$/m<sup>3</sup>) (except sludge disposal cost), adsorption by low cost adsorbents – laterite clay (0. 10 US\$/m<sup>3</sup>), adsorption with Fe oxide coated sand filtration (0. 11 US\$/m<sup>3</sup>), ion exchange (0. 12 US\$/m<sup>3</sup>) and hybrid methods (0. 15-0. 17 US\$/m<sup>3</sup>) were most economical in comparison to other reported technologies (Table 4).

Cost of maintenance is a recurring cost that does make As removal methods less popular in developing countries. Due to complexity and cost barriers, most of the As removal technology may not yield fruitful result, but coupled with social interventions, the targets can be achieved. Thus community participation plays an important role to make the As remediation process sustainable. Therefore, it is important to bring local communities together to achieve the sustainable goals of good health and clean water these are SDG-3 and SDG 6 respectively.

# 6. Arsenic removal Vs SDG 3 and SDG 6 goals

Till date there has been approximately 140 thousand articles are published on As, As contamination, As toxicity, and As removal technologies. But till date the situation of millions of people impacted by As is remain unchanged. Now, the question arises-What is the feasible method to remove As with a practical and social objective of alleviating people suffering due to exposure to this metalloid. UN SDGs have already being formulated by international agency to transition the world to a greener environment friendly future to key goals of SDGs (Total 17 goals) is SDG 3 (Health and Well-being) and SDG-6 (Clean water and Sanitation). The global target is to achieve this by 2030. But with 500 million people, mostly residing in low/middle income countries impacted by As is a barrier to achieve this goals. Hence, this review focuses on the cost effectiveness of prevalent As removal technologies with an overall focus on its practical usability in low/middle income countries.

As per the recent researches, low and middle income nations are mostly affected by As pollution. Unlike other water borne pollutants, being a metaloid, As is difficult to be removed from the aquifer without effective treatment. These treatment methods add cost allocations for the policy makers, making them unpopular. Another option will be to supply drinking water in the Arsenic polluted regions to limit human exposure to geogenic As. Works of Hernandez et al. (2019) suggested the requirement and process for the policy level interventions to install a community based As removal plants for the As impacted rural region.

Works of Roy et al. (2015), highlights the effectiveness of community based water treatment systems in the villages of Malatipur, Barshimul, Sirkapara, Gujastirampur in the Arsenic impacted district of Murshidabad, West Bengal, India. According to Kinniburgh and Smedley (2001), In situ a pilot scale chemical and bacterial oxidation based As treatment plant were installed has worked well. 'Danida arsenic mitigation pilot project' was one of the well-known community level water treatment units for the removal of As from contaminated aquifers in Bangladesh (Jain and Singh, 2012). This method is not efficient for removing high concentrations of As but is capable of reducing the As concentration to permissible levels in Bangladesh. Two primary advantages of using in-situ oxidation techniques are: (i) low cost of installation and no serious sludge-related disposal issue (ii) small-scale facility needed (Paul et al., 2010). Van Halem et al. (2010) observed that the freshly oxidized adsorbed iron was available for co-adsorption of As indicating that the As adsorption is dependent on the amount of adsorbed iron that is oxidized (Luong et al., 2018).

The policy level intervention can focus on community based awareness and aim to improve their access to clean drinking water and sanitation (SDG-6). Identifying polluted ground water zones (wells) and disusing them in favor of non-polluted aquifer can be another alternative for communities dependent on ground water for their primary water needs (Chen et al., 2007). 'Willingness to pay' by effected population for clean drinking water is another criterion that need to be evaluated by policy makers while formulating As removal systems suited for community. For example, people may be willing to pay for availing services pertaining to daily need but may find 'paying for water' unacceptable even if it impacts their health and wellbeing (Orgill et al., 2013). Hence, with research focus continuing on developing As removal technologies, there need to be coordinated policy level interventions with community participation to manage the issue in the larger scale.

#### 7. Conclusions and recommendations

Lower- and middle-income countries impacted by geogenic As pollution in the groundwater need economic and feasible 'As' remediation technologies. Technologies developed follows As-removal efficient is mainly dependent on environmental variables, installation and running cpst, installation charges. The existing technologies need to be efficient, environmentally friendly and cost effective to target achieving the goals of SDG 3 and SDG 6 in low/middle income countries. Most of the Asian countries falling under the low and middle income range are worst affected with elevated As concentration in drinking water. Success stories of arsenic removal at community based units' needs to be studied and implemented in different parts of the globe facing similar issues. Innovating 'easy to use', efficient hybrid technologies with low maintenance cost, low electricity demand, low waste generation is the only way forward in achieving the overall target of delivering 'clean and safe drinking water' to all in order to protect the 'health and wellbeing' of impacted populations. Regular monitoring, community awareness, community participation, research on cost effective As-removal models coupled with policy level intervention to ensure clean water supply to all, should be focused in all As-impacted nations.

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# **Ethical approval**

All authors are informed of the ethical guidelines of the journal and we declare that we have complied with them.

#### Table 4

Cost benefit analysis of popular As remediation process.

Popular process	Treatment Methods	Running cost (US\$/m <sup>3</sup> )	Remarks	
Adsorption	Adsorption on iron oxide coated sand	0. 73	Loss of medium capacity, fouling by inorganic and organics matter and orifice clogging	Chappell et al., 2001; Joshi and Chaundhuri,
	Sorptive filtration using gravel bed containing iron sludge	>0. 71	Presence of divalent cations ( $Ni^{2+}$ , $Co^{2+}$ , $Mg^{2+}$ ) enhances the adsorption. As sludge management is difficult.	1996; Ahmed, 2006 Ali et al. 2001; Kabir and Chowdhury, 2017.
	Ferrous based red mud sludge (FRS)	Material cost 88-118 US\$/ton	Without chemical addition or special care. But sometimes bacteriological contamination may occur	Li et al., 2010
	BUET activated alumina	0. 74–1. 19	Compacted activated alumina column adsorbs arsenic on surface of alumina grains. Not suitable for commercial scale as As contaminated sludge disposal is difficult	Chappell et al. 2001; Ahmed 2001
	Filtration with zero-valent iron (ZVI)	1. 2	90% arsenic removal possible from phosphate and silicate rich water. Regular backwashing of sand filter is required.	Hug and Leupin, 2003
	Fill and draw units	1. 015	Manually mixing of water with oxidant and coagulant at 60 rpm speed for 30 s and overnight sedimentation. Controlled mixing and tank gradient aids floc formation and enhance the removal rate. Not capable to reduce below specific level	Ahmed, 2001, 2006
	Read-F arsenic removal unit	2	Iron removal by sand filter to avoid clogging of resin bed.High selectivity for both As(III) and As(V) under wide range of conditions	Ahmed. 2006; Hanchett et al., 2011
	Laterite as a low-cost adsorbent in a sustainable decentralized filtration system	0. 10	Low cost absorbent. Significantly good efficiency	Nguyen et al., 2020
	Adsorption with iron oxide coated sand filteration	0. 11	Cost effective technology for low concentration of As in water. This process effectively removed methane, ammonia, iron and manganese along with As	Petrusevski et al., 2007
on exchange	An integrated ion-exchange membrane process coupled with Fe co-precipitation	0. 12 (as per rough estimation as per laboratory condition)	The major costs in membrane processes are due to the cost of membranes and due to the need of their replacement	Lopes et al., 2020
everse osmosis	Reverse osmosis unit	0. 52 (Community level unit for inhabitants 20000, while cost increased 0. 45-0. 88 US\$/m <sup>3</sup> for population 5000-50000)	High pressure, high pH and low temperature favoured a more efficient removal of arsenic, whereas an increase in ionic strength reduced the removal of arsenate. Dependency on the skilled personnel. Pretreatment required as preexisting ions interfere the filtration process. Adjusting the pH of raw water can increase the media capacity and lowers the operating costs; however, pH control equipment increasesinvestment costs and the overall operational complex	Abejón et al., 2015
and chemical arsenic in the ac reduction	<i>In situ</i> oxidation of iron and arsenic in the aquifer	0. 076 (except the sludge disposal cost)	It allows formation of iron hydroxide coating on sand grains around the strainer of the well. Recollected water from the aquifer shows around 50–70% reduction of arsenic. Bacteriological contamination may result in some cases	Ahmed, 2006
	The granular ferric hydroxide	0. 043 (except the sludge disposal cost)	Chemical processes include oxidation and precipitation, where each process is carried out in separate operational units and thus increased capital cost associated with the necessary chemicals and equipment	Driehaus, 2002
	Iron-based subsurface arsenic removal (SAR)	0. 06–0. 07	Pilot scale study proved efficient for removal of Arsenic from water in Vietnam	Kurz et al., 2020
ectrocoagulation	Electrocoagulation of As	0. 22–0. 31	This process needs continuous supply of electricity, which is associated with additional initial cost and technical issues. Disposal of toxic sludge is also adds up cost in these processes	Addy, 2010, Addy et al, 2011; Kabir and Chowdhury, 2017
iological remediation	Biological removal and fixation of As		A stable arsenite oxidation efficiency of 99% was achieved, while the removal of total arsenic was 93%. The drawback of biological methods lies firstly, in the predominance of the most toxic As (III), which requires a prior step of oxidation it may be biological oxidation through microbes, algae and fungi. However water contaminated with low concentration of As can be easily detoxify with help of biological treatments	Vega-Hernandez et al., 2021
	Microbial fuel cell	0. 2 (58 USD per Gallon)	This fuel cell was effective for the removal of both inorganic organic contaminates. This technology is mostly suitable for industrial waste.	Tabassum et al. 2021
		$0.\ 17\pm0.\ 02$		Katsoyiannis et al., 2015

(continued on next page)

#### Table 4 (continued)

Popular process	Treatment Methods	Running cost (US\$/m <sup>3</sup> )	Remarks	
	Pre-oxidation with biological method followed by GFH/ adsorption Ozone oxidation followed by Chemical precipitation	$0.\ 15 \pm 0.\ 02$	The cost of adsorbents count is around $0.09 \pm 0.02$ $\epsilon/m3$ ,which is more than 50% of the total operating cost Operational cost including handling of backwash water including thickening and dewatering of the sludge	Katsoyiannis et al., 2015

#### Consent to participate

All authors have participated in the design, planning and writing and validation of results of this work.

#### **Consent for publication**

All authors agree to the publication of this research according to the guidelines of the journal.

#### Availability of data and materials

All data used in this manuscript are available in references.

#### CRediT authorship contribution statement

Akhilesh Kumar Yadav: Data curation, Writing – original draft, Writing – review & editing, Funding acquisition. Harish Kumar Yadav: Data curation, Writing – original draft. Aliya Naz: Methodology, Investigation, Data curation, Writing – review & editing, Visualization. Monika Koul: Writing – review & editing, Visualization. Abhiroop Chowdhury: Conceptualization, Writing – review & editing. Shashank Shekhar: Validation, Resources, Supervision.

#### **Declaration of Competing Interest**

Authors declare that they do not have any conflict of interest.

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