

Contemporary practices in groundwater arsenic remediation and wastewater management in West Bengal, India: a systematic review

Soumyajit Koley*

Department of Civil Engineering, Indian Institute of Technology, Kanpur, Kalyanpur – 208016, UP, India

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Abstract

Arsenic is a notorious carcinogen that is released into subsurface waters by virtue of natural geological and hydrological processes. Such a contaminated water has detrimental effects on human health when it is consumed for drinking. This paper is an effort to address the widely recognized public health crisis of arsenic poisoning in India's state of West Bengal. It is intended to provide an environmentally safe and financially feasible solution for arsenic mitigation that combines both ground water remediation and waste management situations. Treatment of pumped groundwater by Arsenic Treatment Units (ATUs) for providing potable water has been a major arsenic remediation measure in rural villages of West Bengal. But, backwash cleaning of these units generates hazardous sludge rich in toxic arsenic, disposal of which is an environmental concern. Through a field examination, detailed information on the prevalent sludge management practices at remote ATU sites were acquired and reviewed for their suitability. It was ascertained that wrongful dumping of sludge can be avoided by disposal on certain iron-rich sand filters, constructed in-situ using naturally available ingredients. Also, the incorporation of sludge in building materials, especially the clay-bricks, can be an emphatic option for arsenic waste management at commercial scales. This paper thus presents a comprehensive overview of 'in-situ' and 'ex-situ' strategies of arsenic mitigation. An 'in-situ' management aspect utilizes iron-rich sand filters located within the premises of groundwater treatment units for recycling of arsenic-containing wastewater, followed by adsorption and concealment of residual arsenic. An alternative, hazard-free means of stabilizing toxic sludge off-site, with clay-bricks, at a brick manufacturing factory, has been proposed for the 'ex-situ' aspect. Many policy discussions around the globe do not feature arsenic-sludge disposal or stabilization as a topic. Keeping this in view, this review article concludes with innovative approaches to sustainable arsenic remediation and waste management in India.

Keywords

Arsenic, Groundwater, Treatment, Sludge, Sand filter, Bricks.

1. Introduction

Globally, intrusion of toxic heavy metals into groundwater ecosystems has been a topic of serious concern for sustaining public health. Extracting contaminated subsurface waters for consumption as drinking water intake has led to catastrophic debilitations in populations, particularly those in rural areas of developing countries lacking robust municipal water-supply infrastructures [1]. Within this subject matter, mysterious and rampant geochemical release of arsenic in the subsurface waters of India (as depicted in *Figure 1(a)*) has impaired the concerted mitigation efforts of several geologists and environmental science researchers around the world. Arsenic (As) is a notorious carcinogen and its chronic intake causes incurable health problems [2, 3].

It is a metalloid which occurs naturally in different inorganic and organic forms.

The estimated abundance of arsenic in the earth's upper crust is in excess of 1.5 ppm [4, 5]. It is colourless, odourless and tasteless when dissolved in water, and thus cannot be detected easily by simple observation [6]. Factors like water temperatures, pH and salinity, oxidation state, etc. control the speciation, mobilization and fate of arsenic in water [7, 8]. Arsenic goes through a series of modifications by redox reactions, ligand changes and bio-transformations within a pH range of 6.5–8.5 that is typically found in shallow aquifers [9–11].

In the subsurface environments, it occurs largely in its inorganic form, which has been classified as a severe human carcinogen [12, 13]. As little as 70 to 180 ppm of arsenic poisoning in the human body can be fatal,

* Author for correspondence

notably for women [14–16]. Arsenic in drinking water is not allowed to exceed 0.01 mg/L according to the guideline from the World Health Organization (WHO) [17]. Yet, the drinking water in countless rural habitations of South Asia contains arsenic poisoning. Groundwater arsenic contamination in the Bengal Delta Plain, which constitutes Bangladesh and the Indian state of West Bengal, has been a globally known natural catastrophe for over three decades. The geogenic nature of arsenic release in Indo–Gangetic aquifers has been mostly attributed to fluvial sediments originated from the Himalayas [18–20], with pervasion caused due to its mobilization under natural hydro–geological conditions [21–23]. This predicament has been assessed to be the biggest, and the most relentless, case of groundwater arsenic contamination in the planet [24, 25].

India's West Bengal is one of the critically affected regions in this regard. *Figure 1(b)* expresses the extent of this crisis in the region. Six districts in the state, namely Murshidabad, Maldah, Hooghly, North 24 Parganas, Nadia and South 24 Parganas are adversely affected, where over 100 million people consume water drawn from the shallow aquifers bearing gravely high levels of arsenic. The depth of these contaminated aquifers ranges from 15 to 70 m below ground level. Several remedial actions have been put to practice by the state government and various other Non–Governmental Organizations (NGOs) to provide potable water to the people living in these regions. These measures predominantly involve tapping and treating river water (*Figures 2(a)* and *2(b)*) and openly–harvested rainwater (*Figure 3*) for supplying potable water to households via piped–water–supply networks [26].

However, establishment and maintenance of the infrastructure required for such networks are not possible in many remote rural villages due to economic and geographical hurdles. As a result, in these areas, hand–pump–fitted tubewells (*Figure 4*) drawing shallow groundwater remain the primordial source of drinking water, for their easy installation, and availability of the subsurface water for extraction [27, 28]. Hence, arsenic remediation of the extracted groundwater by Arsenic Treatment Units (ATUs) has become indispensable in bringing urgent relief to the arsenic–affected rural habitations [29].

Over the last few years, thousands of ATUs (as in *Figure 5*) have been set up in the arsenic–affected districts, especially in the remote rural villages. Alongside treatment for other major inorganic contaminants (e.g., iron, fluoride, nickel, barium, etc.), the groundwater being mostly pathogen–free in the region, requires much less disinfection compared to that for the river water, in order to become potable (i.e., safe for drinking). But undesirably, the ATUs generate enormous quantities of wastewater that are rich in toxic arsenic. Arsenic is very mobile and can readily leach if the arsenic–bearing sludge is not stabilized [30]. Exhausted treatment media of these units are not significantly hazardous in this context [31].

But the wastewater needs to be quarantined or disposed in an environmentally safe manner to prevent further contamination of soil and groundwater. A field survey of certain notable ATUs was undertaken by the author, and the ongoing waste management practices were noted. Small–scale ATUs that serve about 600 households for supplying potable water are called Arsenic Removal Units (ARUs), whereas large–scale ATUs that serve more households are commonly termed as Arsenic Removal Plants (ARPs) [1]. In a vigorous attempt to present a technical overview of logical viewpoints of theory and practice in arsenic mitigation strategies, with regards to the ARPs and ARUs, this paper explores the sustainability of such policy initiatives in India.

This article is founded on the author's opinion and perspectives of the status quo of groundwater remediation and wastewater management in India, which has been attained via the extensive field investigations of remote rural habitations in the country. Thematically aimed at achieving the global and regional Sustainable Development Goals (SDGs). This review paper primarily incorporates three key issues of sustainable water and wastewater management policies, namely, (1) Water infrastructure and distribution, and environmental stability, (2) Sludge treatment, including processing, storage and management, (3) Resource remediation, innovations and economic recovery, (4) Decentralized water supply and stakeholder awareness, and (5) Social resilience and development. All the acronyms introduced in this article have been listed as an *Appendix I* mentioned at the end of this paper.

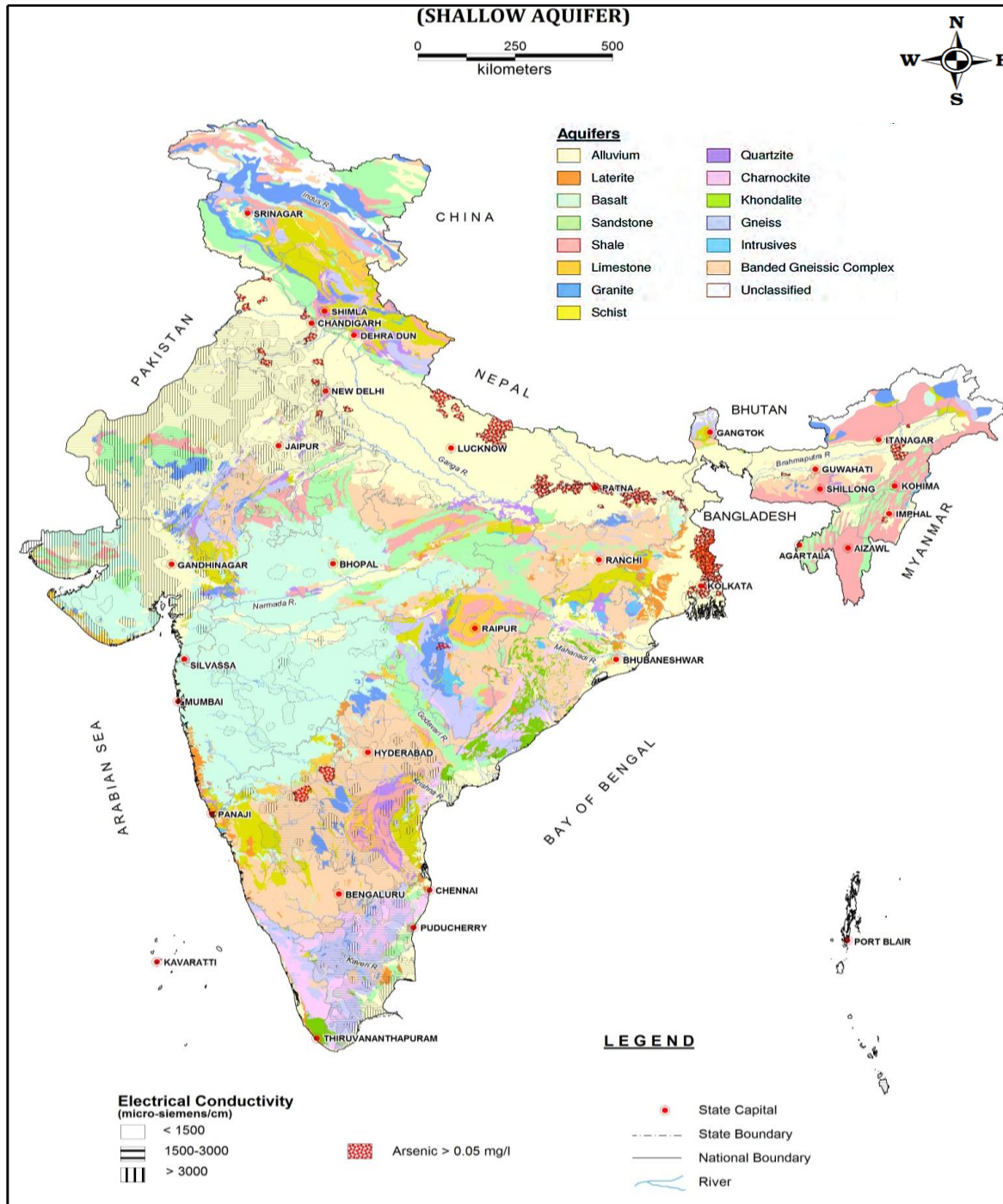


Figure 1(a) Geochemical extent of groundwater arsenic contamination in India (adopted from Koley, 2021 [1] with permission of the copyright owner)

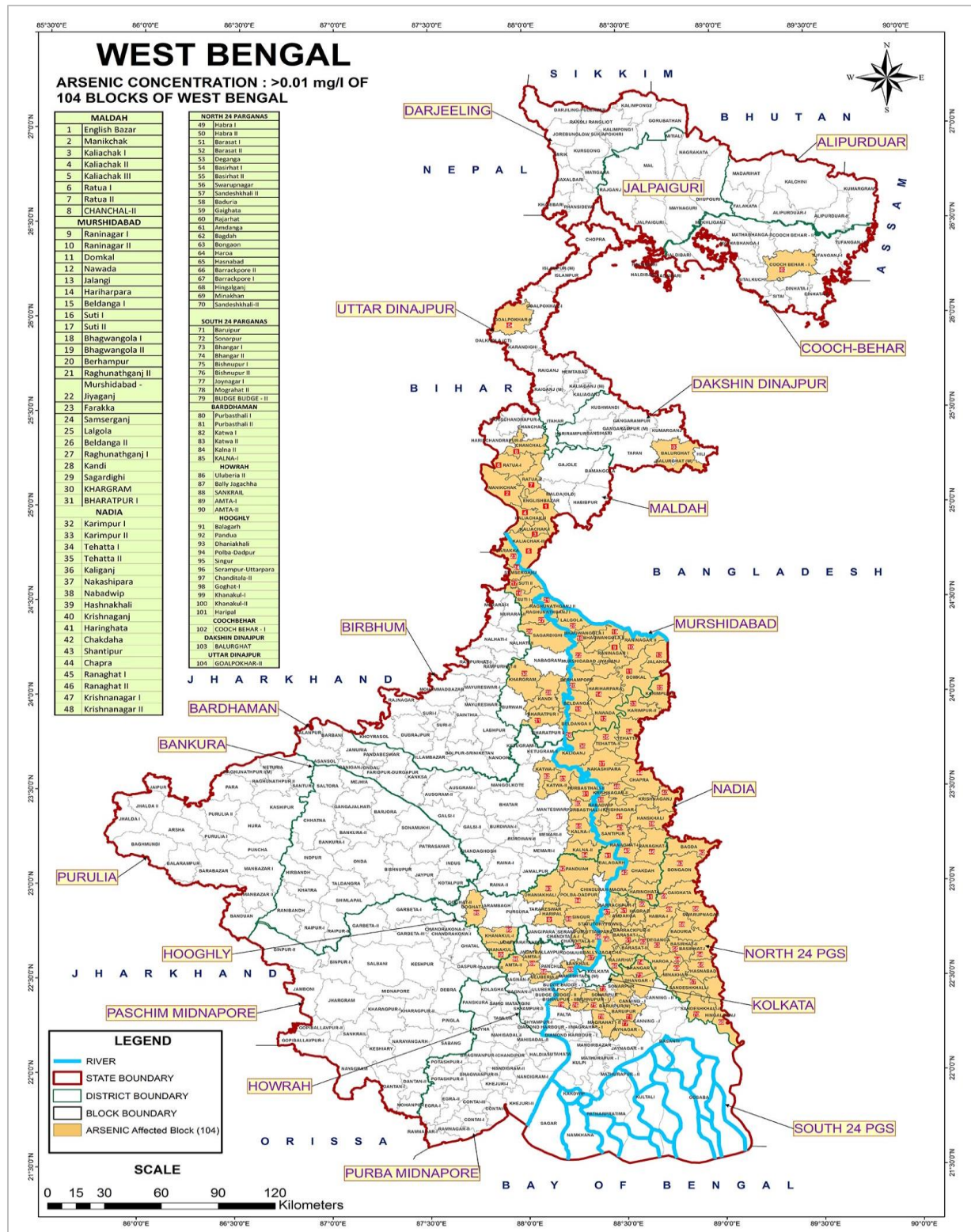


Figure 1(b) Groundwater arsenic contamination situation in the state of West Bengal in India (adopted from Koley, 2021 [1] with permission of the copyright owner)



Figure 2(a) Treatment plant of a mega surface water-based water-supply scheme (Falta–Mathurapur) in district South 24 Parganas, West Bengal. Arsenic-free surface water of the perennial river Ganges is abstracted for Piped–Water–Supply Schemes (PWS)



Figure 2(b) Overhead reservoirs to distribute treated surface water to thousands of households via piped-networks. Administrative names, from left to right: Diamond Harbour I (Zone–2), Diamond Harbour II (Zone–6), Uttar Raipur Budge Budge I



Figure 3 Pond-based rainwater harvesting, water supply scheme in district North 24 Parganas, West Bengal



Figure 4 A typical hand-pump-fitted tubewell in rural West Bengal, that has now been abandoned after detection of arsenic in groundwater



Figure 5 A typical ATU located in doctor para habitation of the Khanjepur village under Tehatta-I block in district nadia, commissioned scheme under Khanjepur water supply scheme

2.Crisis situation in India and remedial measures: An overview of literature

The Bengal Delta plain of India and Bangladesh in Southeast Asia is one of the major arsenic contaminated regions globally. Various hydro-chemical studies carried out in the Ganga–Meghna–Brahmaputra (GMB) river basin reveal the provenience of arsenic to be of geogenic inception [32, 33], that perpetuates and exacerbates spontaneously under natural hydro-geological processes [34, 35]. Oxidation and reduction hypotheses are the most discussed theories to explain the genesis of arsenic mobilization in the subsurface of the GMB basin, with the latter acquiring more scientific acceptance in academia [36–39]. As per the oxidation hypothesis, arsenic is propelled by the oxidation of arsenic-bearing sulfide minerals, like Arsenopyrite (FeAsS), in the shallow aquifers, owing to the drawdown induced by groundwater rampant abstraction from the aquifer [40–43].

In the reduction hypothesis, arsenic is thought to be released through dissolution of Fe/Mn oxy-hydroxides (that are coated on mineral grains in arsenic-bearing sediments) in a reducing environment [44–50]. The groundwater arsenic contamination is widespread in Indian states along the GMB basin, particularly Bihar, Uttar Pradesh, Jharkhand, West Bengal, Assam and Manipur [51–55]. As a result, over 500 million people living in these states have been poisoned by arsenic from drinking water sourced from the aquifers [24, 25, 56, 57].

In the GMB basin, mainly shallow alluvial aquifers (i.e., 15 to 70 meters below ground level) comprise late Quaternary sediments of Holocene age that contain arsenic [19, 21, 58]. In addition to organic matter, the sediment consists of micaceous sand, silts, and clay, which are transported from the Himalayas by the Ganges and its tributaries [25, 59, 60]. The mineralogical composition of these sediments includes mostly feldspar, quartz, kaolinite and illite [18, 49]. *Figure 6* shows the Hydrogeology of the state of West

Bengal in India, where the arsenic calamity is widespread. Since the emergence of this geogenic disaster, numerous attempts have been made to model and estimate the fate of arsenic transport in groundwaters [43, 61–63]. But there are uncertainties involved in the findings [64, 65].

Artificially recharging an arseniferous aquifer by rain water or surface water [66, 67], or injecting oxygenated water [68, 69], have been found to drastically dilute and reduce the As(III) concentrations in the aquifers. But such a contamination regulation technique does not give much success, because of inadequate understanding of all the ecohydrological processes which govern the geochemical behaviour of these aquifer systems [28, 70].

Nevertheless, a multidimensional programme has been initiated for arsenic mitigation in India's West Bengal with projects ranging from establishment of surface water based piped-water-supply schemes (PWS, as shown in *Figure 2(a)* and *Figure 3*) to commissioning of ATUs for supplying potable water to the population in the arsenic-affected zones. Many of these government projects are supervised and funded by international agencies like United Nations Children's Emergency Fund (UNICEF), WHO, World Bank, etc. Resorting to arsenic-free river water of the Ganges for the PWS (i.e. upon suitable chemical and biological treatment) is accepted as the ideal scheme to control the arsenic menace, specifically in the districts where such arrangements can be made possible and beneficial to the society [29].

However, remote rural habitations having no or little access to the surface water based PWS have no other option but to rely on hand-pump-fitted tubewells alone for domestic consumption, particularly in drinking and cooking [71, 72]. Hence, treatment of the extracted groundwater by appropriate ATUs is a vital policy measure for arsenic mitigation in such regions [28, 73].

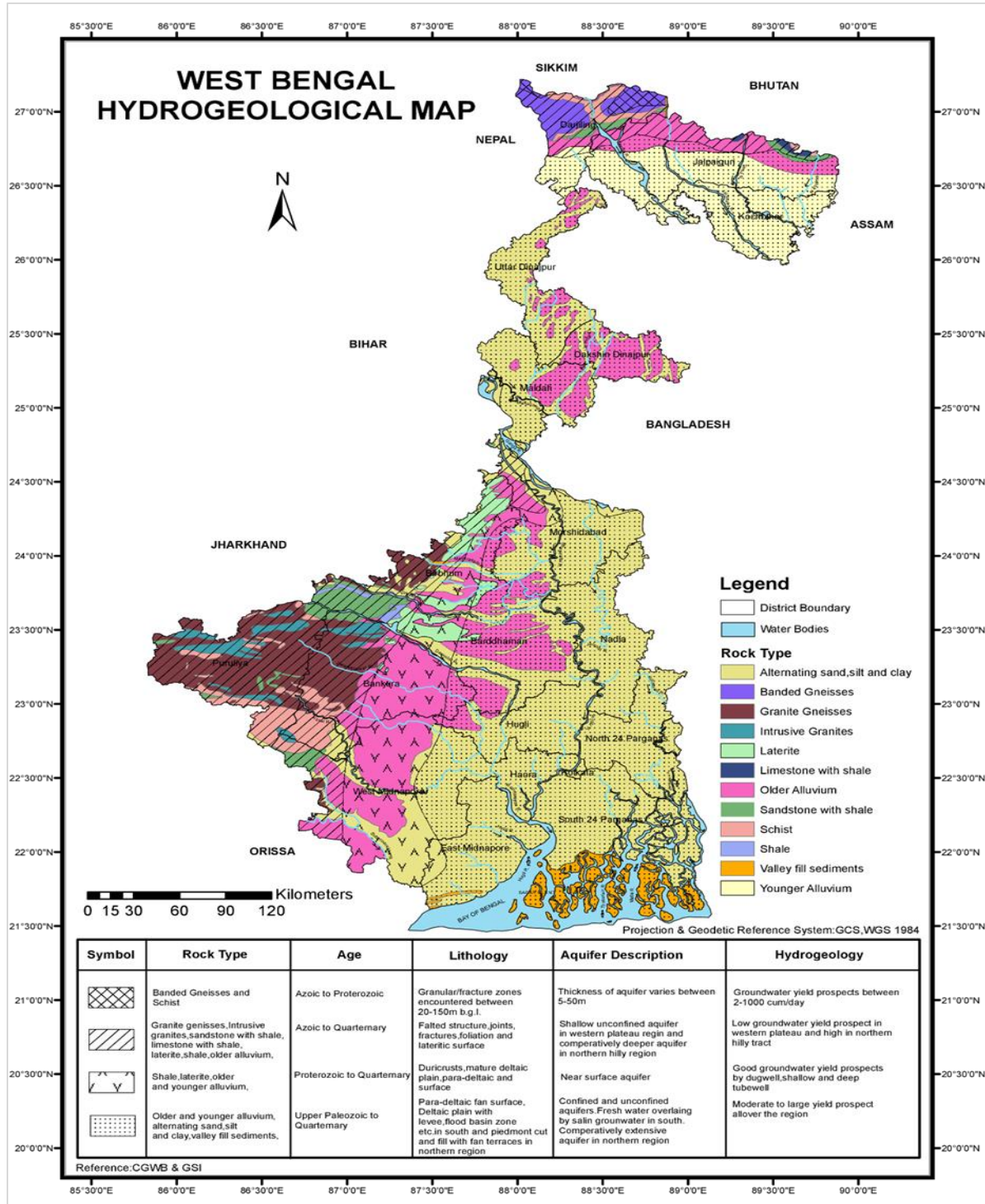


Figure 6 Map showing Hydrogeology of West Bengal, India

3. Arsenic treatment units: costs involved and wastes produced

ATUs are often classified as ARPs (*Figure 7*, operated 5 h/d) or community-based handpump-tubewell-attached ARUs (*Figure 8*, operated 6 h/d). A typical household in the rural regions of India comprises four members, each of whom consume about 5 L of water daily in drinking and cooking [74, 75]. Generally, an ARU serves up to 600 households on a daily basis by providing potable water either on-site, or through water tankers (*Figure 9*). For convenience in procurement by some distant households, water is delivered by means of commonly available 20 L reusable Polyethylene Terephthalate (PET) bottles (*Figure 10*). Usually, mini-trucks like tempos are hired, each having a carrying capacity of 50 PET bottles at minimum, for fast and reliable door-to-door delivery to the distant households. Large-scale ATUs, i.e. the ARPs, serve further more number of households via piped-networks. Operation, maintenance and quality-checks of the units are done by community-clubs composed of trained village personnel employed by government authorities and NGOs. Water tariff to a household ranges from INR 10–20/L in this regard. Social acceptance and sustainability of an ATU depends on several factors such as the arsenic removal rate and efficiency, the water tariff charges, and user-friendliness in the operation and maintenance (O&M) [76]. *Table 1* shows information on costs and water treatment efficacies of certain ATUs (i.e., ARPs and ARUs) which the author identified in field visits to Nadia and North 24 Parganas districts of West Bengal. These ATUs are run on a non-profit basis and have been found to be widely adopted by rural habitations [77]. On an average, the capital and O&M costs of an ATU are INR 67,000 (USD 931) and INR 22,000 (USD 306) respectively. Number of ATUs necessary for installation are dependent mainly upon the number of people in an area that remains at high risk of arsenic exposure via drinking water. For keeping the total arsenic concentration limit at 0.01 mg/L in treating drinking water [31], the ATUs follows the Indian standard specification IS 10500: 2012, while also allowing a maximum limit of 0.05 mg/L in the absence of alternative drinking water sources in the region under consideration [78]. If the maximum permissible limit of arsenic in treated drinking water of the ATUs is strictly enforced to be 0.01 mg/L based on United States Environmental Protection Agency (USEPA's) revised rule, more expenses would be required for the establishment of the upgraded ATUs in this context. This would result in an increased remediation cost of the groundwater [79].

Technologies to treat arsenic in pumped groundwater principally include oxidation, adsorption or coagulation of arsenic, followed by separation of the residue thus produced via filtration [80, 81]. In general, iron, aluminium and their minerals are used as water treatment media in the ATUs, in view of their high co-precipitation and sorption affinity towards arsenic [82, 83]. Some ATUs utilize manganese greensand in combination to enhance the treatment efficiency [84]. The media are typically designed in a way to have ample sorption sites in effectively adsorbing arsenic despite the presence of other competing oxyanions in the groundwater, such as carbonate (CO_3^-), sulphate (SO_4^{2-}), nitrate (NO_3^-), silicate ($\text{H}_2\text{SiO}_4^{2-}$), phosphate (H_2PO_4^-), etc. They also remove other water contaminants like fluoride, lead, barium, chromium, etc. With up to 0.5 mg/L of arsenic in the contaminated feed water, the ATUs often remove more than 90% of the arsenic. However, presence of particulate matter in the feed water and deposition of arsenic-iron precipitates on filter media formed during the treatment of water by adsorption or coagulation, plug the media over time which leads to increasing head loss and reduction in hydraulic capacity of the units. In order to avoid clogging of the media and restore its optimal water treatment efficiency, the system requires periodical cleaning by backwashing. The treatment sludge (of about 1 g/cm³ bulk density) thus removed as waste from these units, either in the form of slurry (from co-precipitation based ATUs, as in *Figure 10*) or brine wastewater (from adsorption or ion exchange based ATUs, as in *Figure 11(a)*), are generally voluminous and encompass high concentrations of arsenic toxicity [85]. Annually, about 550 L of wet sludge is generated from backwash cleaning of an ARU alone [86]. Solid wastes are essentially the spent media, which get enervated over time by arsenic adsorption (*Figures 12(a)* and *12(b)*). Recovery of arsenic by its desorption from the spent media can be achieved with acid and alkaline washing (i.e., by using dilute H_2SO_4 and NaOH respectively) at 20–40 °C temperature range, and it has been noted that low temperatures facilitate the desorption process [87, 88]. This procedure not only helps in recovering the adsorbed arsenic, but also regenerates the media for a further reuse. Exhausted treatment media of adsorption-based units are generally regenerated 3–4 times before being replaced or recharged by new ones [31, 89, 120, 125, 126]. Lately, some pilot studies have been conducted in developing simplistic, robust and affordable arsenic remediation methods using electro-coagulation techniques, which do not require a treatment media or much use of chemicals, and thus produce extremely

low quantities of sludge [90–93]. The techniques make use of electrolytic oxidation of ordinary mild steel electrodes in the contaminated water by passing direct current at a suitable voltage to produce iron oxyhydroxides, which co-precipitate with arsenic to form flocs that can then be separated by filtration. While

efforts are currently underway in developing scalable business models for sustainable implementation of such techniques at electrified villages, successful operation of the existing ATUs and associated waste handling methods continue to remain a difficult task.



Figure 7 A typical ARP located in Char Panpara habitation of Panpara village under Shantipur block in district Nadia, commissioned scheme under Gayespur Water Supply Scheme (Zone-2)



Figure 8 A typical ARU located in the Tajpur habitation of Uttar Tajpur village under Karimpur-I block (GP Pipul Baria) in district Nadia



Figure 9 Water tanker to supply drinking water to distant households



Figure 10 Storage practice for arsenic–sludge by an ARU developed by Central Glass and Ceramic Research Institute, Kolkata, that is located in the Akrapur village of Barasat Municipality, in district North 24 Parganas (adopted from Koley, 2021 [1] with permission of the copyright owner)



Figure 11(a) Wastewater generated by an ARP located in School Para habitation of the Mrigipota village under Chapra block in district Nadia, commissioned scheme under Bahirgachhi Water Supply Scheme (Zone-2)

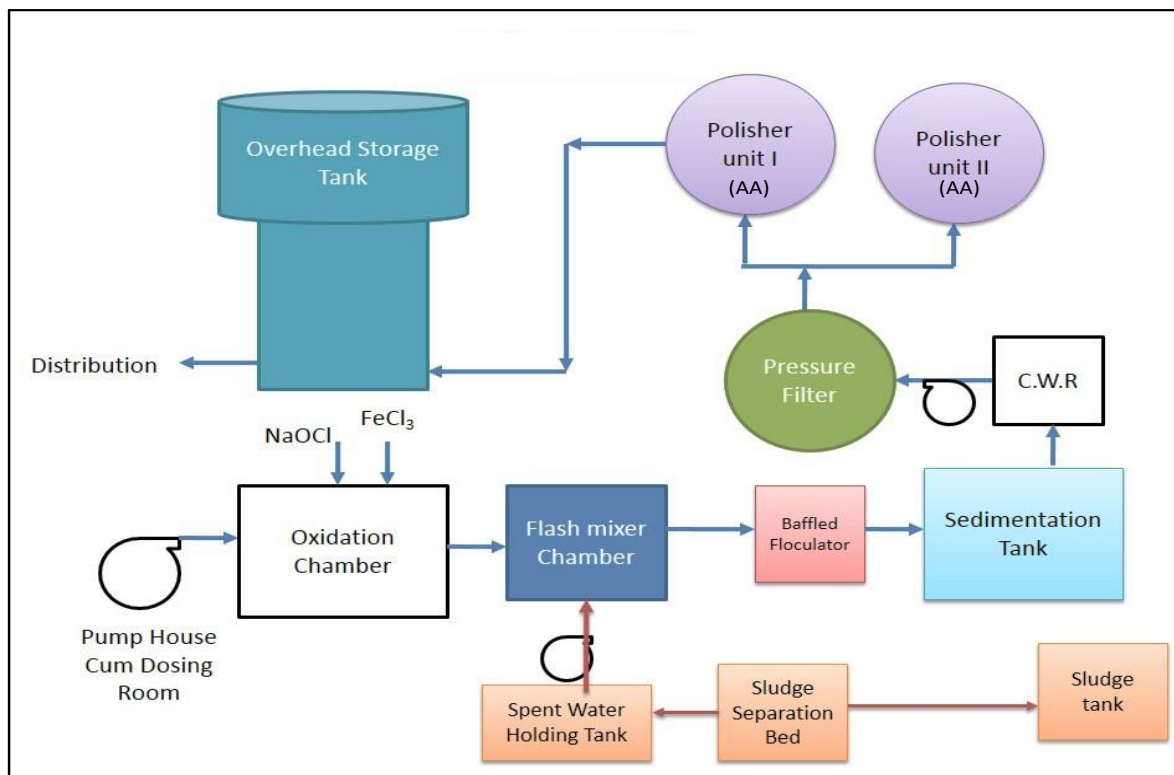


Figure 11(b) Water treatment and wastewater recycling process of an ARP (Note: AA stands for Activated Alumina based water treatment media, and CWR stands for Clear Water Reservoir)



Figure 12(a) Discarded exhausted media of an ARU of Asarpara habitation of Bagula village under Hanskhali block (GP Bagula-I) in district Nadia



Figure 12(b) Discarded exhausted media of an ARU of Mahatpur habitation of the Mahatpur village under Bhagawangola-I block (GP Sudarpur) in district Murshidabad

Table 1 Certain prominent ATUs in West Bengal, India (Data assimilation based on Koley, 2014 [31])

Manufacturer providing Arsenic treatment technology	The location identified on field visits	Village and District		Gram Panchayat ^a	Principle of operation	Filter Media	Media capacity for water treatment	Annual expenditure (in INR)	Efficacy in removal of Arsenic and Iron to below 0.05 mg/L		Total water loss during treatment (% volume of feed water)
									As (%)	Fe (%)	
Public Health Engineering Department (PHED) technology, Govt. of West Bengal (ARP)	Nutannath Bholadanga village, Santipur block, district Nadia	JL No. 4 and GP	Gayeshpur (code 08)	Adsorption	Hematite Activated Alumina (AA) + Manganese green sand	+ 600 – 1,000 L/h	85,000 capitals + 1,680 for O&M	95	95	2	
											Gobardanga town, district N24 Pgs.
AMAL unit developed by Bengal Engineering & Science University (BESU), Shibpur (ARU)	Parpatna village, Deganga block, district N24 Pgs.	JL No. 45 and GP Chakla (code 04)	Adsorption	Activated Alumina (AA)	125 L/h	57,000 capital + 18,500 O&M, including 4,500 for media regeneration	95	90	1–2		
Pal Trockner Pvt. Ltd., Kolkata (ARU)	Ichhapur village, Gaighata block, district N24 Pgs.	JL No. 36 and GP Ichhapur I (code 06)	Adsorption	Granular Ferric Hydroxide (GFH) called AdsorpAs®	450–680 L/h	77,400 capitals + 26,000 per recharging	98	91	1		
All India Institute of Hygiene & Public Health (AIHH & PH), Kolkata (ARU)	Simulpur village, Habra-I block, district N24 Pgs.	JL No. 154 and GP Maslandapur II (code 05)	Co-precipitation assisted microfiltration	Bleaching powder + Calculated Ferric Alum dose	1,000 L/h	51,480 capital + 21,125 O&M, including periodic chemical reagents' charges	95	90	5		
Anir Engineering (AE) Kolkata, in collaboration with ITP GmbH, Germany (ARU)	Faridpur village, Kaliganj block, district Nadia	JL No. 91 and GP Faridpur (code 03)	Adsorption	Granular/slurry Ferric Hydroxide (GFH/SFH)	600–900 L/h	815 capitals + 192 per recharge + 27 for O&M	90	90	1–2		
Central Glass & Ceramic Research Institute (CGCRI), Kolkata	Village Akrampur Talikhola, district N24 Pgs.	Ward No. 4, Municipality	Adsorption followed by cross flow microfiltration	Colloidal adsorbent media + fabricated ceramic cartridge	2,500 L/d	2,96,000 capitals + 4% O&M	96	92	5		

^aA Gram Panchayat (i.e., GP) is basically a cluster of villages for which a particular identification code number is assigned by Zila Parishad (i.e., District Council). JL number refers to the jurisdiction list number allotted by the land reforms department of the state government. For municipalities, a ward number is assigned. Also note, only 1 ARP (i.e., the one in Gobardanga Municipality) currently exists and operates in the district North 24 Parganas. Monetary data are inclusive of taxes, wherever applicable.

4. Toxicity of the arsenic wastes

Arsenic in the wastes of ATUs is found to be mainly sorbed onto amorphous iron and aluminum oxyhydroxides in such wastes [94]. Some of the testing methods which have been devised to check arsenic's leaching potential in such wastes include Extraction Procedure Toxicity Test (EPTox), Toxicity Characteristic Leaching Procedure (TCLP) [95], Waste Extraction Test [96], and Australian Bottle Leaching Procedure [97]. As per USEPA, over 5 mg/L

arsenic content in TCLP leachate qualifies the waste as hazardous [98]. However, the credibility of TCLP in estimating the leaching behavior of contaminants in hazardous waste sites has been questioned many a times [99, 100]. In certain case studies, it has been recognized that arsenic-release in a landfill depends on prevalent redox conditions and their rate of change, the arsenic concentration in the wastes, and chemical composition of landfill leachate which varies from landfill to landfill [101, 102]. Therefore, development of a common or universal method in estimating

arsenic–release in variable environmental conditions remains a challenge. Nevertheless, TCLP is perceived as being good enough to have an approximate estimation of the toxicity characteristics of wastes, and is widely adopted for arsenic–release estimation [103]. *Table 2* shows the recorded values of concentration of arsenic in TCLP extracts of the waste samples. Arsenic contents of the samples were first determined by a semi–quantitative Wavelength Dispersive X–ray Fluorescence (WDXRF) analysis. TCLP extraction

tests were conducted on the samples based on the guidelines and procedure laid out by the USEPA’s Method Number 1311 [104]. It can be seen, that although the values of arsenic concentrations in the TCLP extracts of some wastes are less than the USEPA standards [105], suggesting that they are non–hazardous, the leaching is high with respect to the Indian regulatory standards for effluent discharge from water treatment units [106].

Table 2 Leached contents of arsenic from wastes, estimated by TCLP

Arsenic Waste	Arsenic Treatment Unit (ATU)	Arsenic (mg/L)	content	Arsenic in TCLP extract (mg/L)
Spent AA media	Public Health Engineering Department	860		0.10
Spent GFH media	Anir Engineering	650		0.20
Backwash sludge	Central Glass & Ceramic Research Institute	16,520		15.00

5. Waste management methods

5.1 Prevalent practices

Mixing of arsenic–sludge with livestock waste (e.g., cow–dung and poultry wastes), followed by detaining the mixture in an enclosed biogas plant or open soak–pit, is often a preferred disposal method in remote habitations of India [107, 108], as most of the arsenic was reported to be removed within a time frame of 3–5 months in this process. Validity of bioremediation of arsenic wastes with such organic wastes under aerobic and anaerobic conditions has been evaluated by various studies [109, 110]. It has been ascertained that arsenic is removed in this process mainly due to volatilization as a result of biotic transformation of arsenic species by microbial action, thereby unleashing toxic $\text{As}(\text{CH}_3)_3$ (trimethylarsine) and AsH_3 (arsine) gases, which would have adverse impact to health of villagers living in proximity [111, 112]. Besides, it is a conventional practice in rural areas to obtain a dry cow–dung readily available from the soak pits for cooking purposes. Reportedly, gases liberated by burning of such arsenic containing solids in domestic ovens have indeed led to acute respiratory problems, especially in cases where kitchens are inadequately ventilated [113]. Thus, there are noteworthy health repercussions of using bio–wastes in attempts to reduce the toxicity of arsenic–sludge.

Nonetheless, in an immutable absence of explicit guidelines for waste disposal [114], fixation of a hazardous arsenic waste by stabilization has been assessed to be the most suitable option for long–term sequestration of arsenic in the environment and

inhibition of its leaching [115]. Stabilization of arsenic wastes can be done by commercially or naturally available adsorbents in required proportions [116]. But most of the stabilization methods are not enforceable in a sustainable manner in a remote rural setting of India due to lack of requisite resources, awareness and skills. This is evident in *Table 3*, that briefly shows the literature data on management practices followed by rural communities for disposal of the arsenic–sludge and exhausted media, in the context of some of the ATUs described in *Table 1*. In yesteryears, the arsenic–sludge was simply discharged in nearby landfills, or in municipal–wastewater drains that were mostly channelled towards local ponds and lakes. But it has been examined that repetitive loading of high–arsenic–rich wastes on a particular disposal site could lead to excessive leaching of arsenic, especially under highly alkaline conditions, and presence of arsenic competing bicarbonate (HCO_3^-) and phosphate (H_2PO_4^-) anions in the leachate [101]. Also, the leaching process gets instigated or stimulated in a reducing environment [117]. Indiscriminate open disposal of high volumes of such hazardous wastes would therefore certainly lead to serious re–contamination of underlying aquifers. Moreover, such leachate–impacted groundwater systems may become on–going sources of contamination of nearby surface water bodies as well [118]. It thus becomes important to strictly check such practices and consider alternative methods for hazard–free stabilization of the wastes.

Table 3 Information on current waste disposal and reuse practices at some ATU sites (Data assimilation based on Koley, 2014 [31])

ATU	Practices for arsenic–sludge recovery and disposal	
AMAL	Media restoration process	Monthly backwash of the system
		Regeneration of spending media in a year
		Replacement of the exhausted media by fresh media
	Waste produced	Wet sludge, as the regenerant solution
		Exhausted adsorbent (AA) media, after few times of regeneration
	Waste disposal methods	Clogged sand filter due to build-up of backwash sludge disposed of it
Wet sludge mixed with fly ash, followed by discharge in aerated soak-pit filters		
Exhausted media, crushed and discharged in domestic wastewater drains		
Pal Trockner	Media restoration process	Clogged sand filters are used in cement mortars (up to 5 % by weight)
		Monthly backwash of the system
	Waste produced	Media is non-regenerative, so the media is replaced after exhaustion
		Spent adsorbent (GFH) media
	Waste disposal methods	Wet sludge, as brine wastewater upon media backwash
		Spent media, reused with clay bricks (at 1:6 ratio by weight)
AIIH & PH ^b	Wet sludge is discharged in landfills or domestic wastewater drains	
	Media restoration process	Daily backwash of the system
	Waste produced	Slurry containing coagulated arsenic–iron flocs
	Waste disposal techniques	Slurry is reused with clay–bricks or ready–mix concrete (up to 10 % by volume)
PHED	Media restoration process	Weekly backwash of the system
		Replacement of exhausted media in 2 years (no regeneration is done)
	Waste produced	Spent adsorbent media (comprising of manganese sand, hematite and AA)
		Wet sludge, as brine wastewater upon media backwash
	Waste disposal methods	Spent hematite lumps, reused as raw material in steel plants
Spent activated alumina, stabilized by Portland Cement (at a 1:10 ratio by weight)		
Wet sludge is schematically recycled through an underground sludge separation bed made up of iron–oxide coated bricks		

5.2. Recent developments in waste management

A field investigation of the ATU sites revealed that a lot of progress has been made in the recent while to prevent inadvertent or unethical dumping of arsenic wastewaters. A proper backwash log–book is maintained at most of the sites, which has resulted in curtailing the incorrect disposal practices of arsenic–sludge. This has been possible due to regular inspection by government personnel, and increased awareness and training among the communities handling the ATUs. The ARPs have been potent in curtailing the arsenic–sludge disposal issues by way of ‘in–situ’ stabilization of the waste [31, 119] (as shown earlier in *Figure 11(a)*). Customarily, systemic arrangements are made in an ARP’s premises to recycle the arsenic–rich water generated upon periodic backwashes of the water treatment media (as depicted schematically in *Figure 11(b)*). Separately constructed sludge separation bed (built within the premises itself) adsorbs the arsenic in the recycled water, thereby enabling it to be retreated and further recycled. In order to adsorb and contain the extra arsenic in

residual water, the bed can be formed by placing layers of iron-rich river sand on clay-brick chips, while ensuring adequate head space. Consequently, not only is the wastewater from the ARP treated and recycled, but the sludge that is generated as a result, also stays concealed within the unit’s premises. However, on a negative note, ARPs distinctly require skilled manpower to set up, along with high technical expertise and frequent supervision for the intricate O&M, which are not feasible in many remote rural areas having marginal and sparse populations. ARPs are thus difficult to facilitate in remote rural areas lacking vital infrastructural support. Such ATUs lose social acceptability over time, if they are not monitored regularly and operated proficiently [77, 120]. Besides, there always remains a possibility of accidental outflow of arsenic from the sludge separation bed during extreme weather events. On the contrary, the ARUs have been readily and widely accepted by rural households in India as they are easy to manage and operate [77, 107]. But, ARUs generate copious volumes of arsenic–rich wastewater upon

backwash cleaning of their media. Safe disposal of this toxic wastewater is a burdensome challenge, and therefore it is stored and kept isolated [31]. Unlike the ARPs, not all ARUs have provisions in their premises for 'in-situ' stabilization of the sludge, due to which the sludge is quarantined in drums or in the commonly available PET jars. In some places, where ARUs are installed or run by NGOs and private agencies, these wastes are sold to local manufacturers of value-added construction materials, on a profit or non-profit basis,

for its stabilization (*Figure 13*). But, such an 'ex-situ' arsenic stabilization practice is yet to be adopted by government bodies on a wide-scale so as to ultimately dispose the intense volumes of toxic wastewater generated from various ARUs annually. Nevertheless, following sections comprehensively reveal the information on the current waste management policies.



Figure 13 A construction site in Gangarampur Municipality in district Dakshin Dinajpur, utilizing value-added construction materials incorporating waste products such as agricultural residues and municipal wastewaters

5.2.1. In-situ waste stabilization

In all ARP sites, and in some ARU sites, the wastewater (i.e., the wet sludge) is kept and monitored in the same premise, by disposal on 'in-situ' constructed open sand filters as sludge separation beds [31,121,122]. However, at most ARU sites where the backwash sludge typically has high turbidity, indicating high toxicity, the sludge is isolated by storage in enclosed drums or the 20 L PET jars (*Figure 10*). Nonetheless, a sand filter is constructed by digging the ground in a cylindrical or rectangular section to a depth of about 1.5 m below surface followed by adding layers of fine and coarse-grained sand in the pit. The sides and bottom of the pit are sealed by lining with cement mortar or a mixture of

lime and clay, so as to prevent accidental leaching and transport of arsenic and iron due to run-off or infiltration of water during rainfalls. Care is also taken to cover the filters in such situations to prevent arsenic transport by erosion. The upper sand layers are coated with natural iron-oxides to enhance the stabilization process. Marine sediments deposited in the banks of the river Ganges which are rich in goethite and hematite [123], are used as the iron-oxide mineral coatings. By keeping the filters open during the day, adequate aeration is provided for the rapid stabilization of the toxic species of arsenic. As the excess water evaporates, oxidized iron-arsenic complexes remain safely encapsulated in the filter. Disposal of arsenic-sludge of iron-rich sand matrices

have been found to be suitable for cost-effective in-situ inhibition of arsenic leaching under aerobic conditions [124, 125]. At certain sites, brick lining is done at the sides and no sealing is made at the bottom to allow the filtered arsenic-free water to seep into the ground [126]. No adverse impact on groundwater systems in the area has been reported in such cases. The filters are replaced annually with fresh ones. Clogged sand filters are sent to government laboratories where attempts are being made to recover raw arsenic and iron [31]. Decontamination of sand filters can be done on-site by washing with Ethylene Diamine Tetra-acetic acid (EDTA), followed by recovery of arsenic and iron from the resulting chelant solution by silica gel-boned Solid Phase Extraction (SPE) [127, 128]. Regeneration of the SPE system is done by backwashing with HNO_3 and the consequent treated chelant solution obtained as a result can be utilized again for recycle as the EDTA. The washed sand filters can be reused or openly discarded on aerated environments. Annual cost of regeneration of a sand filter by this procedure of acidic and caustic wash constitutes 80% of an ATU's operation and maintenance (i.e., O&M) cost. Favorably, with regards to volume and arsenic-leaching behavior, the solid wastes and spent regenerant solutions are generally found to be non-hazardous (i.e., arsenic in TCLP leachate is much less than 5 mg/L, as evident in *Table 2*), which dismisses any plausible cause of concern for their disposal.

In a few cases, sludge is mixed with fly ash obtained from ash-pond-dump sites of coal-based thermal power plants located in the rural areas, followed by disposal in aerated soak-pits. This seems alright in theory as fly ash has been found to be very effective in entrapping arsenic due to its high contents of aluminium and ferric oxides that readily adsorb the arsenic [129, 130]. Besides, the recovery of raw arsenic from fly ash can be easily done with acid washing using dilute H_2SO_4 [131]. However, all batch experiments recommending such disposal methods have been done with arsenic salts ordinarily available in labs, and none of the experiments used actual sludge (of field conditions) generated from ATUs. Also, no credible information exists on adsorption, desorption and leaching characteristics of other toxic heavy metals like lead, copper, chromium, nickel, cadmium, etc. which accompanies arsenic in the sludge. Moreover, the fly ash itself in the country has been found to contain extremely high concentrations of such pollutants [132]. It also contains trace levels of arsenic and has the potential to contaminate water resources [133]. Hence, the use of hazardous industrial

wastes in stabilization or co-disposal of arsenic wastes should be discouraged, as they do not provide practical solutions with regards to sustainable arsenic waste management.

5.2.2. Ex-situ waste stabilization

With regards to certain ARUs, stabilization of arsenic-bearing sludge by incorporation into construction materials like cement mortars and concrete is quite common (as evident in *Table 3*), which gives a scope for recycle or reuse of the waste on a commercial scale. Laboratory investigations by batch and column adsorption studies have shown that arsenic is successfully stabilized in ordinary Portland cement due to adsorption with calcium hydroxide [134–136]. It has been identified that mixing up to 40% of sludge by volume to cement-sand mortar or concrete does not cause significant arsenic leaching [137–139]. For many years, rigorous static and semi-dynamic leaching experiments have been carried out for studying the longstanding leaching behavior of arsenic from the Portland Cement stabilized matrices, and also in combination with other cementitious binders like lime, fly ash, etc. [140–143]. It has been determined that arsenic leaching increases in alkaline conditions due to decalcification or dissolving of ettringite gels as a result of carbonation of calcium-bearing minerals. As cement is typically used in mortars and ready-mix concrete that are mostly exposed to alkaline conditions (e.g., during rainfall), high arsenic leaching from the stabilized product can therefore be anticipated. Moreover, no conclusive information is available on the physical strength characteristics of the stabilized products, which restricts the reuse of arsenic with such materials on a commercial scale.

But, interestingly, stabilization of arsenic-sludge by means of infusion with modular clay-bricks (as in *Figure 14*) have been supported by numerous studies to be a superior waste management method, which presents another scope for its commercial reuse. In moist clay-moulds prepared for the brick making, initial arsenic adsorption occurs due to electrostatic bonding between positively charged arsenic-bound iron hydroxide solid phases with negatively charged colloidal clay phases [144]. At optimum sludge-mix proportions, the arsenic is effectively encapsulated in the vitrified clay-moulds while burning [86, 145]. Efforts have also been made to determine the best ratio of sludge and firing temperature for maintaining brick durability and strength as a consequence of the addition of sludge [146–148]. It has been concluded that superior or adequate quality sludge-mixed bricks are obtained in the typical firing temperature range of 900–1300°C itself. In 1st-class quality Indian modular

bricks, the wet sludge is permanently encapsulated, by mixing it homogeneously at 1–10% by volume. In neutral and alkaline exposure environments, the arsenic-laden bricks can be safely used as ornamental bricks and as a building material in masonry, with the sludge addition of up to 6% and 4% by weight, respectively. In every case, arsenic encapsulation is > 98%, with the stabilized brick being non-hazardous in terms of arsenic leaching phenomenon (i.e., arsenic in TCLP leachate < 0.02 mg/L). Unfortunately, this is not always true in case of acidic exposure environments (e.g., urban heat islands, industrial setups, places with high intensity of acid rains, etc.) in which arsenic leaching increases due to dissolution of excess iron hydroxides which fail to find available charges for bonding with clay [144, 145].

Nevertheless, such bricks can be safely used in load or non-load bearing interior walls of residential structures, though such a technique should not be used for stabilizing wastes like a spending media waste containing high amounts of alumina and silica. Excess silica (> 70% w/w) makes a brick brittle and weak on burning, and excess alumina (> 40% w/w) produces cracks in brick on drying and makes the bricks refractory [149, 150]. Also, commercialization of mass scale stabilization of sludge with bricks would result in higher economic benefits to the government and the society, which would help to further invest in arsenic mitigation schemes in the long run. Such benefits have been projected to be of the order of millions of USD, that is worth investing in rural development projects of India [31]. With the booming brick production market in India's alluvial Gangetic plain, it shall be worthwhile to execute a waste management project of arsenic wastes' valorization with bricks, based on the location, character and quantity of the waste to be stabilized. Ex-situ stabilization, through which arsenic-sludge gets permanently restrained in the environment, will help in avoiding long-term storage of the waste and subsequent disposal or processing elsewhere that presents a perpetual risk of accidental leaching of arsenic. In this way, no extra effort, so as to dispose

the sludge on-site or off-site of an ARU, is required for the waste management, which saves valuable time and money. Therefore, an ex-situ waste management project shall help the government in achieving robust and sustainable arsenic mitigation (i.e., groundwater remediation and waste management, together) in India.

5.2.3. Integrating water treatment and waste stabilization as a dynamic system

Figure 15 schematically summarizes both the 'in-situ' and 'ex-situ' plan of action, of arsenic mitigation, in two separate scenarios using a dynamic modeling technique. The in-situ management approach mandates arsenic-bearing wastewater to be recycled through iron-rich sand filters located on-site, before adsorption and the sequestration of residual arsenic. The ex-situ management approach is envisaged to make use of clay-bricks at a brick manufacturing factory to stabilize the toxic sludge off-site and in a hazard-free manner. As evident in the diagram, the dynamic model can also perform a cost-benefit analysis in order to calculate the benefit-to-cost ratio for the primary stakeholders (i.e., government and society) in the two scenarios, so as to determine the best mitigation strategy (i.e., water treatment in combination with associated sludge-stabilization aspect) that is deemed to be most sustainable for implementation in the study area. The calculations go clockwise from sectors 1 to 4 in the *Figure 15*, via feedback loops. Solid and dotted lines with arrow heads manifest positive and negative feedbacks, respectively, between connected entities. Highlighted parameters portray static or predefined values which initiate the feedback response, while others denote the dynamic ones. The model thus created is also designed to estimate the consequent welfare gains (in the form of health benefits) to the society as well. Though slightly complex in nature, the formulation of this analytical model is quite robust, and besides assimilating numerous parameters of fiscal data, it also integrates several other factors of demographic changes and the establishment of various water-supply schemes



Figure 14 Clay-bricks being used for construction of an ARU in rural West Bengal (location Gosaichar Purbapara habitation of the Gosaichar village under Ranaghat-I block in district Nadia, GP Nawpara Masund)

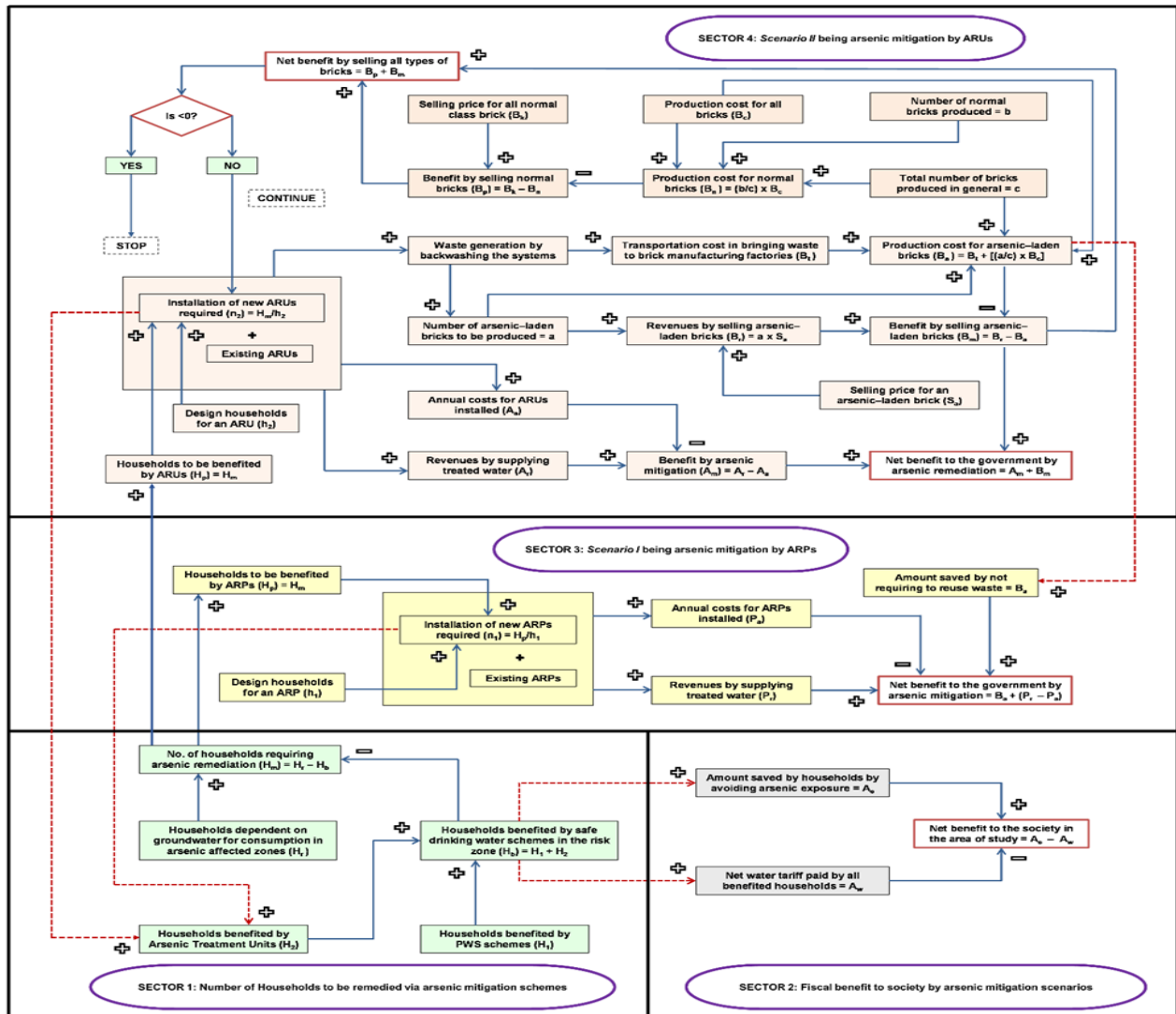


Figure 15 Schematic outline describing the arsenic mitigation strategies as an integrated dynamic system (Note: + sign refers to positive feedback, while – sign denotes negative feedback between the connected components)

6. Conclusion

Geogenic arsenic contamination of shallow aquifers is a critical issue challenging the socio-environmental progress of the developing world. The situation is worse in a populous nation like India, where the majority of the rural population relies heavily on groundwater for drinking water. For over three decades, millions of households in India's state called West Bengal have been at the tragic end of this public health crisis. Remedial measures involving piped-water-supply of treated surface water and harvested rainwater are ultimate ways for attaining sustainable arsenic mitigation in such a region. However, these schemes can cater to only a limited population due to logistic and financial constraints. In such a case, the establishment of ATUs, that treat extracted groundwater for providing safe drinking water, would remain as the principal remedy for arsenic remediation in the remote rural habitations of the country. A field investigation of the ATUs revealed that management of toxic and voluminous wastewater generated from these units upon backwash cleaning is necessary to achieve effective arsenic mitigation. Inadvertent and careless disposal of the waste in landfills or municipal drains can lead to recontamination of underlying aquifers, thereby hindering the sustainability of the ATUs. In other words, all the diligent efforts of the ATUs in eliminating arsenic from groundwater become futile in lowering the overall arsenic toxicity of the environment. Therefore, stabilization of the wastewater is vital to encapsulate the arsenic. Practical utilization of industrial wastes or metallurgical by-products for stabilization or co-disposal of arsenic-sludge is also a challenge due to the highly hazardous nature of these substances towards the environment. Yet, in the absence of a robust infrastructure for waste disposal in rural societies, the waste management can be done either by in-situ quarantine and monitoring of the sludge, or by ex-situ recycling or reuse of the waste by stabilization with common building materials such as clay-bricks. A meticulous overview of literature in this regard shows that incorporation of the arsenic-sludge into clay-bricks are a promising solution for the sustainable management of wastewaters in the arsenic mitigation projects.

This review paper proposes a novel solution to the disastrous arsenic problem in West Bengal, India, which addresses both groundwater remediation and waste management issues simultaneously in an environmentally safe and economically sound manner. With this article, a new viewpoint has been offered on the ground realities of arsenic mitigation policies in environmental management projects. Certain ATUs

called ARPs have provisions within their premises to stabilize the toxic wastewater in-situ. However, ARPs necessitate enormous area and skilled manpower for investiture and operation, which may not be expedient for a long haul, particularly in a rural setting with sparse populace. On the contrary, several small-scale ATUs called ARUs have been greatly accepted by rural households due to convenience in their use and sustenance. But the ARUs produce high volumes of the hazardous arsenic-sludge, that is normally kept stored on-site, which ultimately require potent stabilization for safe disposal. As establishment of ARPs is not quite reasonable in rustic habitations due to socio-economic barriers, arsenic remediation plan incorporating installation of ARUs and subsequent off-site stabilization of arsenic wastes with bricks must be accomplished for permanent encapsulation of arsenic in the local environment. Ex-situ stabilization, where most of the waste will be utilized to make bricks, while also getting secluded from the environment, will not only assist in preventing long-term storage of the toxic waste, but also prohibit its subsequent disposal elsewhere that has an accompanied risk of accidental leaching of arsenic into the environment. In a post COVID-19 economy, commercial benefits accrued by governmental implementation of such projects shall contribute in poverty alleviation and modernization of the rustic communities as well [151-154]. Policy measures that incorporate such methods should be encouraged and enforced.

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Conflicts of interest

The author declares no conflict of interest. This paper is an original, independent, and a standalone work that adheres to all best practices for publishing. The author's M.Tech thesis has been the inspiration behind writing this review article. No financial or institutional support is involved in any form in preparing this paper.

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Soumyajit Koley has been a Consultant for the Water Quality Monitoring and Surveillance Programme of the Government of West Bengal in India. His research expertise lies in formulating sustainable resource management policies as effective

mitigation strategies to combat climate change and outbreaks of infectious diseases. He earned a Bachelor of Technology degree in Civil Engineering in the year 2012, from the esteemed Hooghly Engineering & Technology College (affiliated to West Bengal University of Technology) of the state of West Bengal. Subsequently, in the year 2014, he also attained a Master of Technology degree in Civil Engineering (specialization: Engineering Geosciences) from the prestigious Indian Institute of Technology, Kanpur of India. He is an avid reader of Open Access publications and believes that scientific knowledge should be accessible by anyone desirous of achieving academic glory and wisdom. As a nationalist, he wishes to see India become self-reliant in international scholarly publishing, in the coming decades.

Email: samkoley5@gmail.com

Appendix I

S.No.	Abbreviation	Description
1	AA	Activated Alumina
2	AE	ANIR Engineering
3	AIIH & PH	All India Institute of Hygiene & Public Health
4	ARP	Arsenic Removal Plant
5	ARU	Arsenic Removal Unit
6	ATU	Arsenic Treatment Unit
7	BESU	Bengal Engineering and Science University
8	CGCRI	Central Glass and Ceramic Research Institute
9	CWR	Clear Water Reservoir
10	EDTA	Ethylene Diamine Tetra-Acetic Acid
11	EPT _{ox}	Extraction Procedure Toxicity Test Method
12	GFH	Granular Ferric Hydroxide
13	GMB	Ganga-Meghna-Brahmaputra
14	GP	Gram Panchayat
15	INR	Indian Rupee
16	ITP GmbH	It is a private company based in Germany, that manufactures industrial goods and equipment
17	NGO	Non-Governmental Organization
18	O&M	Operation and Maintenance
19	PET	Polyethylene Terephthalate
20	PHED	Public Health Engineering Department
21	PWS	Piped-Water-Supply Schemes
22	SDG	Sustainable Development Goals
23	SFH	Slurry Ferric Hydroxide
24	TCLP	Toxicity Characteristic Leaching Procedure
25	UNICEF	United Nations Children's Emergency Fund
26	USD	United States Dollar
27	USEPA	United States Environmental Protection Agency
28	WDXRF	Wavelength Dispersive X-ray Fluorescence
29	WHO	World Health Organization