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ABSTRACT: The rapid growth in the use of fertilizers and pesticides in agriculture, excessive extraction of groundwater, and rise in the number of industries with inefficient waste disposal system have been some of the key factors in degradation of groundwater quality during the past years. Although groundwater is considered as a valuable natural resource, the quality control of this resource has systematically failed in India. Irrespective of rural or urban locations, the average sulphate contamination of groundwater in India has reached 90 to 150 mg/L. Such a borderline contamination concentration poses threat both to livelihood and to economy. In addition, the negative health effects of sulphate-contaminated drinking water can range from dermatitis to lung problems and skin cancer. The biostrategic manipulation of groundwater discussed in this article involves sulphate-reducing bacteria used in addition to a 3-step procedure involving constitutive aeration, filtration, and shock chlorination. With earlier use of a similar strategy in the United States and Europe proven to be beneficial, we propose a combinatorial and economical approach for processing of groundwater for removal of sulphur contamination, which still largely remains unnoticed and neglected.

KEYWORDS: Sulphate, groundwater, sulphur-reducing bacteria, filtration, shock chlorination, constitutive aeration

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Introduction

Groundwater is one of the most pervasive, economical, and high utility-oriented resource for both residential and commercial purposes. This layer marks the zone of saturation and is enveloped by a superficial vadose zone, occurring due to capillary dimensions of water trapped in these interconnected voids.[1] The availability of groundwater refers to water-saturated zones found below the earth's surface occurring due to the filling up of the interconnected void of rocks. However, such an enormous availability and ease of extraction of groundwater have also faced an uncontrolled utilization of groundwater over decades.[1–3] Despite the social and geographic demographic dividends among rural and urban areas, there has been a noticeable rise in groundwater consumerism in both the social landscapes.[1] The common patterns of such exploitations range from high scale of agricultural and domestic water requirements in rural areas to intensive water usage by several industries and urban households. In view of these conditions, our focus in current years has shifted from managing groundwater quantity to ensuring higher standards of groundwater quality. At international level, the World Health Organization (WHO) has framed quality standards, which has set parameters for measuring permissible concentrations of common water contaminants.[2]

Broadly, the common pollutants reported from various parts of the world can be grouped as physical, chemical, biological, and radioactive solids or soluble chemicals.[3] The regulation and measurement of these contaminants in India are performed by a large-scale survey consisting of sampling and measurement from various potential locations done by Central Ground Water Board (CGWB).[4] Like the rest of the world, India has witnessed a heightened change in the physical and chemical properties of groundwater owing to changing natural conditions in the form of local geology, climate pattern change, and frequency of rainfall changes taking place over decades. Apart from these natural deviations, a developing economy like India has witnessed excessive utilization of low-cost fertilizers and pesticides, noncentralized sewerage system, open dumping of toxic industrial effluents, and over-abstraction of water. This has led to percolation of contaminants into subsurface zone of saturation in aquifers.[3] Also, the microbial population residing in these voids mediate several biogeochemical reactions leading to alterations of rock composition and release of free minerals into these aquifers.[5] The latest groundwater reports of CGWB, in 2007, identify an average presence of high salinity with an increase in the levels of nitrates, chlorides, sulphates, and suspended particulates.[2,4] Degradation of groundwater can be noticed in distance of primary water source, indicating a secondary mechanism of diffusion and percolation of contamination nearby the industrial and densely populated areas.[3] Thus, not only the primary sources of industries but also the nearby places can be categorized as problem areas for understanding groundwater contamination.[3]

Sulphur as Contaminant

Much of the present research and interest have been aimed to understand arsenic, nitrates, fluorites, chlorides, manganese, iron, copper, chromium, and faecal coliforms and dissolved solids as sources of contamination.[3,5,6] Cases such as sulphur contaminant removal in Minnesota, USA, and Eurogypsum have proved that significant global attention and interventions have proved to be successful in combating sulphur contamination. However, over the past few decades, sulphur contamination of groundwater in India is largely neglected and remains underestimated. In addition, our knowledge of sulphate contamination in the present timescale has been limited to numbers only, and no effort on the elimination of sulphate contamination has been visible. Thus, in this article, we try to escalate the contemporary issue of sulphur contamination of groundwater in India.

Sulphate, one of the major constituents of groundwater, can range from 1.00 to 1000 mg/L depending on the geographic and economic scenario of the place.[1,5] The WHO level of sulphate in groundwater has been set as 250 mg/L and has been permitted up to a higher limit of 400 mg/L. In parallel to the global standards set by the WHO, the Indian standards for groundwater quality monitored by CGWB, India, define sulphate contamination as above 150 mg/L and permissible up to 400 mg/L.[3,5,7] Most of the case studies reported in the subsequent sections of this article will highlight sulphate contamination above the permissible limit in industrial areas, areas nearby large agricultural farms, as well as areas of dense population with nondiscriminatory disposal practices. The CGWB has identified 14 principal aquifers consisting of 42 major subaquifers for identifying sulphate contamination in India.[4,6] The estimation of sulphur prevalence in the metropolitan cities, such as Delhi, Mumbai, and Chennai, has been in the following range: air, 50 to 60 g/m^3 [$SO4^{2-}$] (NAPM, India, 2010)[8]; dissolved water, 400 mg/L (BIS, 2009)[9]; and surface water, 50 to 60 mg/L (BIS, 2009).[10] This is of particular importance because the metropolitan cities in India reflect the situation of densely populated cities with high economic and industrial activities.[8] In addition, the villages nearby or in the neighbouring location of these cities have moderate to high agricultural activities.[8]

Isotopic analysis of sulphur has been instrumental in determining the source of contamination.[4,5,11] In New Zealand, such a pilot study of water from rivers and lakes has allowed distinction of source of contamination as either natural or anthropogenic. The study reported that sedimentary rocks had relatively higher contribution of sulphur [$SO4^{2-}$], 34S isotopic variant of sulphate, than that of industries and households, and this net process of mixing of different water sources leads to net levels of sulphur contaminants.[5,11]

SOx (variable sulphur oxide) adds to the atmospheric levels of sulphur and has been identified as an indirect to low level of threat to lives.[12] Water containing SOx tastes bitter and in severe cases, because of its characteristic laxative property, results in dehydration. This foul taste and associated medical conditions serves as an identification of potential sulphur-contaminated groundwater and needs immediate isolation and treatment.[12] Hydrogen sulphide is another such secondary contaminant, which has a distinct odour and corrosive property and is a threat to life forms.[13] Several such sources of sulphate, sulphite, and sulphide have mounted the overall levels of sulphur in water sources in most Indian cities, varying from 90 to 150 mg/L.[3,5]

Identification of Problem Areas

In this article, we focus on 8 densely populated metropolitan or major cities and 12 potential problem areas to understand the contamination of groundwater in India. The identification of these areas is based on the reports of CGWB, India, and groundwater quality data have been extracted from these reports of 2007. As there has been no such report of systematic groundwater quality assessment after 2007, our study is based on the reports of 2007. With the rising trend of groundwater contamination, several research institutes, academia, and nongovernmental organizations have come forward to develop voluminous quality measurement data and are working towards a strategic technology development that can be implemented with the resources available in India.[14] This article reviews this elusive field of sulphate contamination of groundwater and the potential utilization of sulphur-reducing bacteria in elimination of this contamination.

Sulphate Contamination in India

Sulphur contamination in India has been mainly seen in populous cities, such as Delhi, Mumbai, Chennai, Kolkata, Ahmedabad, and the mining and industrial cities, such as Dhanbad, Singrauli, and Angul-Talcher district (Figure 1).[4,14] Coal, fertilizers, chemical industries, cement factories, explosive factories, ancillary units, refineries, petrochemicals, thermal power plants, aluminium plants, and gypsum building materials are major sources of sulphate contamination in India.[5,6,11]

In particular, coal and gypsum in states such as Andhra Pradesh, Rajasthan, Gujarat, Kerala, Maharashtra, Odisha, West Bengal, and Tamil Nadu (Figure 1) contribute primarily to the widespread contamination of groundwater in India.[11] The 4 metropolitan cities (Delhi, Mumbai, Kolkata, and Chennai) have been identified to have higher sulphate contamination in groundwater which is beyond the prescribed limit of 150 to 200 mg/L.[11] Also, few other densely populated and industrial cities such as Agra (Uttar Pradesh), Coimbatore (Tamil Nadu), Chennai (Tamil Nadu), and Madurai (Tamil

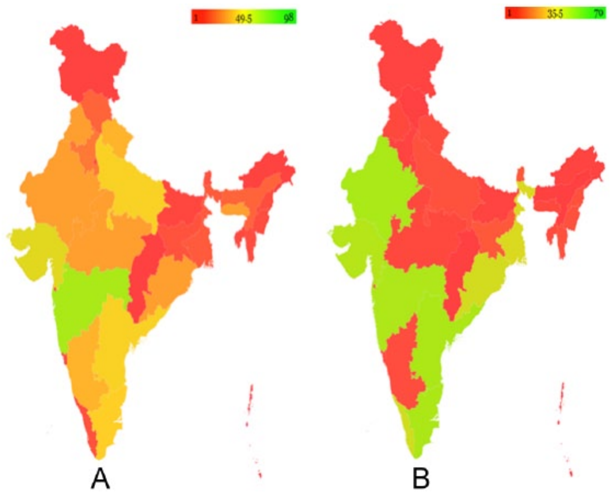

Figure 1. (A) and (B) Plot of groundwater pollution in Indian states. (A) Relative plot of groundwater contamination: Andhra Pradesh, Gujarat, Haryana, Jharkhand, Madhya Pradesh, Meghalaya, Odisha, Uttar Pradesh, Karnataka, and Tamil Nadu are among the leading states with groundwater contamination. (B) Relative plot of states where sulphate contamination is a major source of groundwater pollution: Andhra Pradesh, Gujarat, Kerala, Maharashtra, Odisha, Tamil Nadu, and West Bengal. The maps were generated using the online editable maps available at https://gramener.com/indiamap/.

Nadu) (Figure 2) have been reported to suffer from higher burden of sulphates in groundwater.[4,15] Furthermore, open disposal of toxic chemicals and industrial wastes containing sulphate into water sources and on the soil surface has percolated and diffused down nearby places to cause sulphate contamination of groundwater in these above-mentioned cities.[15]

Other areas of India, identified to be moderate to mid-densely populated cities, and industrial belts have also been identified for existing contamination of groundwater. These have been classified into different problem areas and are listed as Angul-Talcher area (Talcher district, Odisha), Singrauli area (Madhya Pradesh), Chembur area (Greater Bombay, Maharashtra), Tarapur area (Boisar district, Maharashtra), and Digboi (Tinsukia district, Assam). Especially, Singrauli area has an objectionable contamination of sulphate (2338 mg/L) (Table 1) in groundwater and must be addressed with urgent and stringent rules so as to overcome health disaster in this upcoming energy capital of India (Figures 1 and 2).[11,16]

However, the Chembur problem area had a wide range of sulphate in groundwater from 18 to 239 mg/L (Table 1). The average sulphate concentration in entire Chembur area has been within the prescribed limit, but local high concentrations cannot be denied.[11] Tarapur, Talcher, and Digboi have sulphate concentrations at a range higher than the prescribed limit of 150 to 200 mg/L (Table 1). The case of Damodar River Basin has also been unnoticed for its high level of sulphate contamination in groundwater.[4] The Damodar River Basin houses approximately 46% coal reserves of India and thus involves extensive mining and industrial activities leading to rapid hydrogeochemical reactions between the rocks and water as well as percolation of surface contaminants.[11,16]

Sulphate Contamination Outside India

Massive construction activities in developed countries have also increased the environmental burden of sulphate-producing gypsum. To regulate and control the exploitation of environment, either in the form of air or water, the European forum called Eurogypsum has been voicing for closed loop recycling.[8] This sustainable approach of 'closed loop recycling' involves using gypsum again and again as the chemical composition of the raw material in plasterboards and blocks always remains the same.[8] The Eurogypsum also voices the concept of 'alpha diversity' at the global scenario which promotes maintenance of higher biodiversity. All the countries of European Union are stakeholders and accountable to the legislations of this forum and decide their respective diversity indices.[8] A major notable concept of this Eurogypsum policy involves maximizing developmental activities with minimal environmental side effects. Integration of biodiversity protection with recycling of gypsum gives a strong basis for ensuring sustainable and eco-friendly gypsum and its raw materials.

Sichuan Basin in China is another such example of high sulphate contamination. One of the most heavily populated industrial belts in China has experienced an uncontrolled degradation of groundwater resources originating from natural and anthropogenic sources in varied forms discussed above.[13,17] The observed values of sulphate found in the core of the basin with several industries and dense population have been 250 to 300 mg/L, and this has been observed to increase downstream of this basin (350-400 mg/L).[17]

Sulphur-Reducing Bacteria

Sulphate-reducing bacteria such as *Desulfovibrio desulfuricans* have been used for fixing the elevated sulphate levels in water body for more than a decade by now.[16] Bacterial sulphate reduction is a naturally occurring process that proceeds only in the absence of oxygen and in the presence of sufficient organic carbon and sulphate giving rise to elemental sulphur, water, and carbon dioxide (equation (1)). The subsequent reactions lead to the production of hydrogen sulphide in the aqueous conditions:

$$SO_4^{2-} + \text{Organic matter} \xrightarrow{\text{anaerobic bacteria}} S^{2-} + H_2O + CO_2$$

Equation (1) shows the metabolic reaction of sulphur-reducing bacteria.

However, the terminal end product generated, ie, hydrogen sulphide (H_2S), is known to be a negative stress on the environment. This is because of the release of sulphuric acids, growth of iron (Fe)-rich bacteria, and formation of pyrites.[8,16] So, we propose to remodel the use of sulphur-reducing bacteria in a tripartite stepwise manner, remove sulphate contamination,

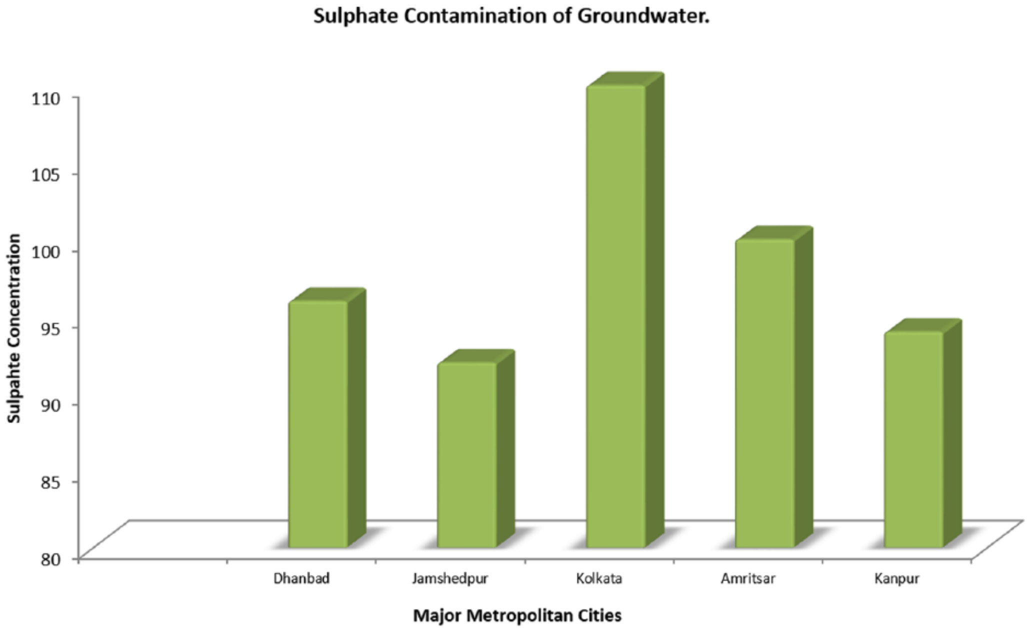

Figure 2. Levels of sulphate contamination in certain major metropolitan cities in India.

Table 1. Number of polluted water bodies state-wise, Central Ground Water Board reports.

NAME OF THE STATE	TOTAL NO. OF WATER BODIES	RIVERS	LAKES/TANKS/DRAIN/ETC
Andhra Pradesh	8	3	5
Assam	2	2	N/A
Delhi	1	1	N/A
Jharkhand	1	1	N/A
Gujarat	10	9	1
Haryana	3	2	1
Himachal Pradesh	2	1	1
Karnataka	6	4	2
Madhya Pradesh	5	4	1
Maharashtra	15	15	N/A
Meghalaya	5	5	1
Odisha	5	5	N/A
Punjab	3	3	N/A
Rajasthan	3	3	N/A
Tamil Nadu	7	7	N/A
Sikkim	1	1	N/A
Uttar Pradesh	8	8	N/A
West Bengal	1	1	N/A
Total	86	71	15

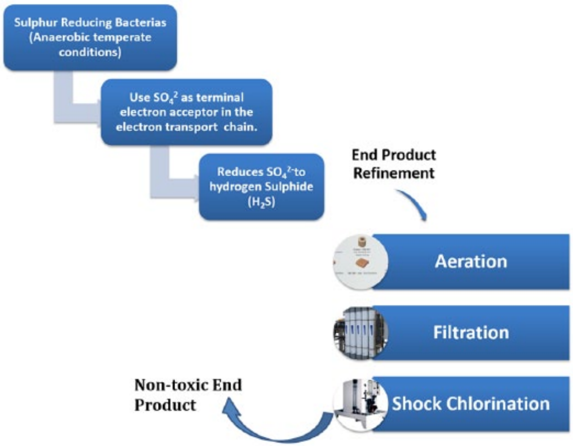

Figure 3. The proposed model of biostrategic sulphur elimination with tripartite end product refinement.

and further convert the noxious H_2S to environmentally non-harmful substances.

Sulphur-Reducing Bacteria in India: Appraising the Contemporary Scenario

The major distinctive property of these sulphur-reducing bacteria is their ability to use sulphur as the terminal electron acceptor in the process of respiration.[12] This facilitates the process of environmental sulphur sequestration and removal of toxic sulphur from ecosystem, eg, from the groundwater as in our case. However, to appraise the situation of sulphur-reducing bacteria in India, it is first needed to locate this diverse group of prokaryotes in different parts of India and try to understand the applicability of these microorganisms in groundwater treatment.

Several indigenous sulphur-rich barite mines found in Kadapa district of Andhra Pradesh, Baula chromite mines of Odisha,[4] and specifically in the southwest marine ecosystem of India contain many of these sulphur-reducing bacteria.[13] Especially, on the south-east coast of India at the Pichavaram mangrove forests, an enormous reserve of microorganisms is present.[14] So, identification and conserved growth of these microorganisms will facilitate in ensuring general supply of these bacteria for setting up huge groundwater treatment plant. This also aims to meet the demand for these sulphur-reducing bacteria from the indigenous sulphur-rich mines and belts along with the marine ecosystem of South India.[15]

One of the reasons for their abundant availability in these regions is the conducive environment ranging from saline to nonsaline ecosystem with a near-neutral pH, a very low redox potential of 100 mV, and a suitable range of temperature.[17] Existing literature reviews suggest that the importance of the sulphur-reducing bacteria in removal of toxic sulphur has been studied in India since the last decade, but the mobilization of this concept into setting up groundwater treatment plants has mostly been neglected.[18] Some of these literature reviews mention the mechanisms adopted by these bacteria which involve the reduction of metal sulphides along with the formation of their insoluble forms, removal of toxic sulphur from waste water, and finally recycling the water.[18]

In a study by Venugopal and colleagues (2000),[19] there has been mention of these indigenously found prokaryotes as bioremediating agents and thus supporting our proposed application for treatment of groundwater in India. With an existing scientific know-how and demonstrated success of similar plans in the United States and Europe, we are hopeful of an equally successful outcome of this cost-effective and indigenous approach for groundwater decontamination.[20]

Considering this richness of Indian ecosystem with the sulphur-reducing bacteria and their biological role in reducing metal sulphides and using sulphur as terminal electron acceptors make them our unique candidates for treating the groundwater in India. In addition, the enormous load of massive reserve of Indian groundwater can be effectively addressed with the low cost and largely available varieties of sulphur-reducing bacteria and their further enrichment cultures in laboratories. This will generate active and large-scale groundwater treatment plant based on sulphur-reducing bacteria and the consecutive process of constitutive aeration, filtration, and shock chlorination as discussed in the following sections to get rid of the toxic byproduct of these bacterial metabolism. This substantiates our overarching hypothesis and implementation suggestions for groundwater treatment in India.

Biostrategic Utilization of Sulphur-Reducing Bacteria

The proposed model of sulphur elimination from groundwater involves initial anaerobic degradation using sulphur-reducing bacteria followed by subsequent tripartite refinement of hydrogen sulphide (toxic byproduct of bacterial metabolism) to non-toxic end products (Figure 3). We prefer labelling our model of sulphur removal from groundwater as 'biostrategic manipulation' because the core of this model involves using microorganisms and their metabolism properties to convert sulphur and different complexes of sulphates into hydrogen sulphide. This is further strategically eliminated by 3-step cleanup procedure involving constitutive aeration, filtration, and shock chlorination. To provide theoretical support and infrastructural feasibility, we have assessed this model based on similar models deployed at sulphur elimination plants at Minnesota, USA, that had shown 60% result efficiency.[18]

As discussed earlier, the primary step is to establish large enriched cultures of selected sulphur-reducing bacteria and use them in a limited oxygen or anaerobic conditions for treatment of groundwater:

> *Step 1. Constitutive aeration* (for less than 10 mg/L $[\mathrm{SO4}^{2-}]$ in groundwater). This process involves both anaerobic and aerobic (with oxygen) treatments of aeration (Figures 3 and 4) to a level much higher than a primary treatment system,

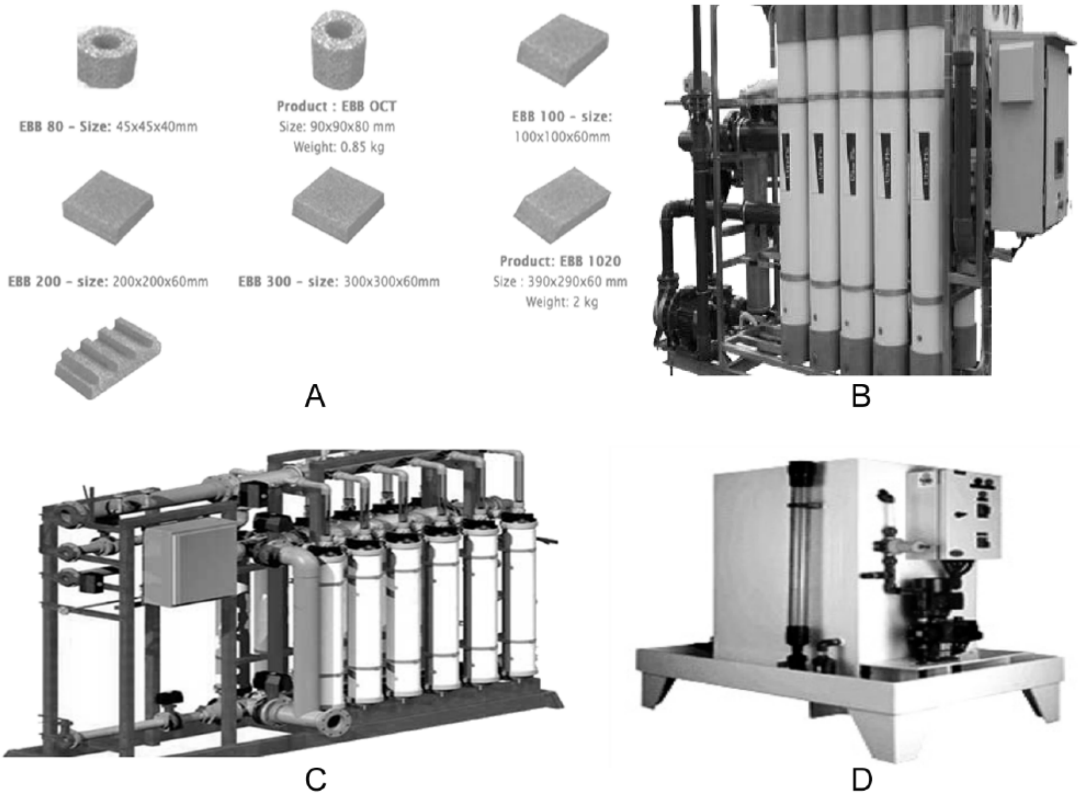

Figure 4. Schematic representation of constitutive aeration, filtration, and shock chlorination apparatus and kits: (A) constitutive aeration kit called 'Eco-bio blocks' of different dimensions, (B) ultrafiltration filtration apparatus, (C) nanofiltration filtration apparatus, and (D) shock chlorination apparatus.

resulting in effluent that is suitable for garden (excluding fruit and vegetables) and landscape irrigation.[21,22] In the conventional practices, constitutive aeration broadly encompasses carbon dioxide-based reduction, ie, decarbonation, a subsequent oxidation of iron and manganese found in groundwater or well water, and a step of stripping the ammonia and hydrogen sulphide from the groundwater.[21] This method of air stripping as a part of constitutive aeration is critical in our model as our compound in focus for elimination is hydrogen sulphide.[21] In a typical aeration of water, the waterfall aerator (commercially available prototype) uses spray nozzles to break the water into small droplets or a thin film of water. This helps to eliminate the effect of air contact. The requirement of a proper air-water contact is because of the fact that it helps in the removal of unwanted gases from the water.[22] However, in the air diffusion method (another commercially available prototype), air is diffused into a collecting vessel which contains countercurrent flowing water and thus leads to small air bubbles.[21]

Step 2. Filtration (for less than 40 mg/L [$SO4^{2-}$] in groundwater). Filtration can be of 2 types: ultrafiltration and nanofiltration. Ultrafiltration is a pressure-driven particle size–dependent purification process in which water and low-molecular-weight substances permeate a membrane, whereas particles, colloids, and macromolecules are filtered.[19] Such a method of filtration has been effective in the removal of colloids, proteins, bacteria, pyrogens, and other organic molecules larger than 0.01 µm in size (Figure 4).[19,23] In contrary, nanofiltration (Figure 4) is essentially a liquid phase one because it separates a range of inorganic and organic substances from solution in a liquid mainly, but by no means entirely, water.[23]

Step 3. Shock Chlorination (for less than 75 mg/L [$SO4^{2-}$] in groundwater). Shock chlorination is performed by mixing a large amount of sodium hypochlorite (in form of a powder or a liquid), such as chlorine bleach, in water (Figures 3 and 4).[22] After shock chlorination, water should be used when sodium hypochlorite falls to or below 3 ppm. In cases where this decline in sodium hypochlorite does not occur sufficiently, sodium thiosulphate can be used as an effective neutralizer.[23]

The proposed strategy even finds suitability with available resources and infrastructure in India. Indigenous companies, such as EBTEC, SR Environ Pvt. Ltd, BioCity, and Delphis Eco, perform the commercial manufacturing of these devices for filtering groundwater contamination.[22]

Conclusions

In the coming years, increase in industrialization, urbanization, and agricultural expansion will accompany the growth in population, economy, and social necessities of the people. This will undoubtedly create pressure on the existing natural resources, such as groundwater, which has indispensable role for various water-demanding activities. The contemporary scenarios of sulphate contamination in various states and regions of India have been identified with similar cases existing at international level. The idea of bringing the natural sulphur-reducing bacteria for clearance of sulphate contamination and subsequent processing involving aeration, filtration, and shock chlorination will answer to the future sustainability of water resources. India, as a country, has a pressing need for fostering its food security programme, and uninterrupted supply of quality groundwater is pivotal in this direction. The common feature of sulphate contamination reported in 4 metropolitan cities and 5 defined problem areas has contaminants originating from both natural and anthropogenic sources. In the long run, developing centralized sewerage system, organized dumping of toxic chemical wastes and limit on the capacity of groundwater abstraction can be promising in mitigating sulphate contamination of groundwater.

Author Contributions

Conceived and designed the paper : SS. Analyzed the data and articles: SS, RKS, CK,RN,KM and KB. Wrote the first draft of the manuscript: SS and RSK. Made critical revisions and approved final version: SS,RSK,CK,RN,KM and KB. All authors reviewed and approved of the final manuscript.

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