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WATER POLLUTION (G TOOR AND L NGHIEM, SECTION EDITORS)

Challenges and Solutions for Sustainable Groundwater Usage: Pollution Control and Integrated Management

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Abstract

Purpose of Review This paper aims to critically review the current status of groundwater usage from the point of view of pollutant control and integrated management.

Recent Findings This paper has shown that sustainable efforts must be encouraged to minimize the arsenic content from all the possible sources before entering the groundwater system. Excessive nitrate and pesticide utilization must be significantly reduced for a sustainable environment. Although various in situ remediation technologies are possible to remove some contaminants in the groundwater, the future concern is how it can be carried out in accordance with environmental sustainable goal such as the implementation of in situ bioremediation and bioelectroremediation which provide a cheaper and greener solution compared to physical and chemical approaches. To develop a successful integrated management for a sustainable groundwater usage in the future, conjunctive water management is recommended as it involves the management of ground and surface water resources to enhance security of water supply and environmental sustainability.

Summary This paper critically reviews the current state of knowledge concerning groundwater usage from the point of view of pollutant control and integrated management. Information presented in this paper is highly useful for the management of groundwater not only in the quality point of view but also in the sustainable quantity for future development.

Keywords Groundwater · Pollution control · Integrated management · Conjunctive water management

Introduction

Recently, a considerable literature has grown up around the theme of sustainability particularly in the area of exploration of natural resources such as agriculture, energy, and water resources. In general, the concept of sustainability is defined as a process and mechanism to achieve sustainable development [1] or it can also be described as a strategy with primary purpose of preventing the depletion of natural resources [2]. In the context of agriculture, this concept is related to the production of long-term crops and livestock with the minimal negative effects to the environment [3]. For the energy, sustainability can be associated with the provision of adequate, reliable, and affordable energy, in conformity with social and environmental requirements [4]. Moreover, the sustainability in water resources particularly for groundwater is the concept adopted for the development and use of groundwater for meeting current and future demands without causing unacceptable consequences to environment, economic, and social. Based on the aforementioned definition, it is likely unrealistic to have a single sustainability concept for every country or region because of the limitations imposed by the social and demographic issues, existing technology, and environmental aspects. Therefore, it is critical for each country to develop applicable and realistic sustainability concept but with a global objective in mind.

Groundwater is an alternative water resource that is relatively clean compared to surface waters. Some countries have used the groundwater for agricultural irrigation and drinking water such as in India, Malaysia, and Indonesia [5–10].

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Among these, India is considered as the largest groundwater user globally. India have used the groundwater for agricultural irrigation and drinking water supply by 60% and 85% of the total demand, respectively [11••]. However, the groundwater resources are currently under high pressure [12••]. This might be due to some factors such as the unrestricted development and increasing pressure for irrigated agriculture and water demand. The problem in supplying reliable water resources is predicted to increase, and it can cause the rise of competition to exploit the groundwater resources, leading to a decline in groundwater levels [12••]. For instance, over the past 50 years, the use of groundwater for agricultural irrigation in India has been increased rapidly with an output of more reliable crop yields and reduces the poverty [13]. However, this causes a negative output which is the falling of groundwater levels and rapid depletion on groundwater. Thus, management plan on the groundwater is important for sustainable development.

Currently, there has been a huge concern about the presence of contaminants in the groundwater because the groundwater is susceptible to some organic and inorganic contaminants [14]. Several contaminants such as natural and synthetic dyes, organic compounds, and inorganic compounds have been detected in the groundwater possibly coming from natural and anthropogenic activities. Several reliable in situ remediation methods such as physical, chemical, and biological have been applied in full scale, but they are still continuously revised and evaluated to improve their efficiency and performance. Although these methods are capable of degrading some groundwater contaminants, the current concern is how it can be achieved in a greener and cheaper manner in accordance with environmental sustainability goal. Therefore, biological remediation methods such as bioaugmentation, biostimulation, and bioelectroremediation become more popular since they provide a quite similar removal performance or higher in some cases better than chemical or physical remediations.

The wise use of groundwater resources has been a concern as the dropping of water table of the groundwater can lead to an increase in the cost of energy for pumping and additional surface supplies of groundwater probably become limited and more expensive [15]. Groundwater management is an essential element to integrate the science in groundwater with the decision of water management [16•, 17]. In the groundwater management, monitoring is a significant action for the water management. Groundwater monitoring can help to track the changes in the groundwater levels and help to provide a better understanding of groundwater condition for the management and make a suitable policy choice, which means a proper regulation to make sure the sustainability of groundwater. The information of the groundwater contaminants can also be identified, and the measurement of the contamination level can be measured more comprehensively and accurately. Through this information, the groundwater management can be developed effectively in order to maintain the water quality of the aquifer and manage the potential effects on ecosystems and public health. In addition, flow pattern of the groundwater is also needed to be managed to prevent the groundwater contaminants or the saltwater intrusion flowing towards the pumping station, which then potentially damage the operation.

Groundwater monitoring is useful to assess the possible negative effects of climate change such as drought. Groundwater management includes several complicated problems with the involvement of multiple stakeholders with different types of decision making for competing goals [18]. Managing groundwater involves the interaction with social, economic, and ecological components, but it is, currently, in an uncertainty range due to the lack of knowledge about the groundwater levels or condition [18]. Groundwater resources usage, particularly in some developing countries, can be probably higher than the surface water usage, which leads to the further increase in drilling and pumping of groundwater [19]. The usage of groundwater exceeding the recharge over periods can cause the declining on the water table and affect the nature groundwater discharge [20]. There might be some negative or harmful impacts towards the ecosystem [19].

This review paper focuses on critical discussion of the challenges and potential solutions in the management of groundwater. This paper focuses on two aspects, which are in pollution control and integrated management. The review predominantly elucidates the different types of groundwater pollutants and their effects towards a sustainable environment. An overview regarding in situ pollution remediation methods is presented along with the challenges and potential solutions for implementing the methods at a wider scale. It is then followed by the discussion on the challenges and potential solutions for the implementation of integrated management from the perspective of stakeholders and human setting. A case study on the exploration of groundwater in Malaysia is provided as an example. Future outlook and recommendation for pollution control and integrated management are also discussed.

Groundwater Contamination

Groundwater is an alternative water resource and has been widely explored in several countries. For instance, the population of China is increasing continuously, and the use of groundwater is about 90% for providing drinking, industrial, and irrigation [21]. However, it was found that the depletion of groundwater in the arid areas has occurred due to the unsustainable anthropogenic activities in China which leads to the threatening in domestic or industrial water supplies and crop yields [22]. The overexploitation of groundwater also causes several issues which include ecological damage and land subsidence that can affect human health. It was also found that more than 60% of the groundwater in China has been found to be contaminated due to poor groundwater management [23]. Through a national water quality survey, the study observed that several harmful contaminants have been detected. Iron, manganese, and arsenic are the most common contaminants detected in the groundwater [21]. Several activities including industry and agriculture are the main factors for the formation of groundwater contamination. Groundwater systems are considered as a part of human nature system. Thus, a comprehensive groundwater system assessment framework has to be established in order to maintain the stability of groundwater and minimize the depletion of groundwater [24]. For instance, a comprehensive evaluation of groundwater resources based on driving forces, pressures, states, impacts, responses (DPSIR) framework was carried out in Iran [25]. The study successfully identified the main driving forces influencing groundwater resources. In addition, extensive groundwater overdraft and decreased aquifer recharge because of reduced rainfall intensity, destruction of vegetation cover, and land use change were identified as the main pressures on groundwater exploration. Currently, a combination of DPSIR framework with Tobit model can be an alternative and has been capable for the evaluation of the performance of water environment in China [26]. The study employed the water environment performance index as the assessment criterion. In general, it was observed that the water environment performance in the study location showed a trend of decline and low performance. Moreover, the Tobit model can be used for analyzing the influence of indicators on water environment performance and can be an alternative to be implemented in other regions as a framework.

Industrial activity also produces pollutants becoming groundwater contamination. For example, production of fossil fuel for the purpose of thermal power generation can be a source of groundwater contaminant. The leakage of petroleum hydrocarbon occurs during the transportation, and the contaminants retaining on the ground surface can be then transferred to the groundwater system [19]. The uncontrolled hazardous waste sites can also lead to the contamination of groundwater as the hazardous materials can flow through the soil and enter the groundwater system [21, 27]. The contaminants have an impact on the distribution of groundwater resources for long term and cause several impacts to the wildlife and human health.

Arsenic has been widely found as a contaminant in the groundwater over 70 countries [28]. This results in severe health hazard as it affected about 150 million people around the world [28]. Around 110 million of those 150 million people are from Southeast Asia and South Asia countries including Pakistan, Bangladesh, China, Laos, Cambodia, India, Vietnam, Nepal, and Myanmar. These severe health hazards have been widely spread through the drinking water supplies from the groundwater [28]. In prolong period, it threatens the people's health and livelihood and eventually causes fatality. As the arsenic substances travel through the soil, in the end, they end up in the groundwater straight to water supply and also accumulate at crops which people can consume daily [28]. There are common established arsenic metabolism models as presented in Fig. 1 [29–33]. They include classical model, model proposed by Hayakawa et al. [32], and model proposed by Naranmandura et al. [33]. The classical model proposed that the alternate reductive and oxidative addition of the methyl group is the main mechanism for the conversion from arsenic to methylated arsenical. Hayakawa et al. [32] assumed that metabolism of arsenic can be facilitated by arsenic-glutathione complex while Naranmandura et al. [33] believed that its metabolism can occur when it binds with soluble and insoluble proteins as shown in Fig. 1. In the future, sustainable efforts must be made in order to minimize the arsenic content from all the possible sources [28].

In natural waters, arsenic can be present in the form of arsenite (As(III)), arsenate (As(V)), monomethylarsonic acid

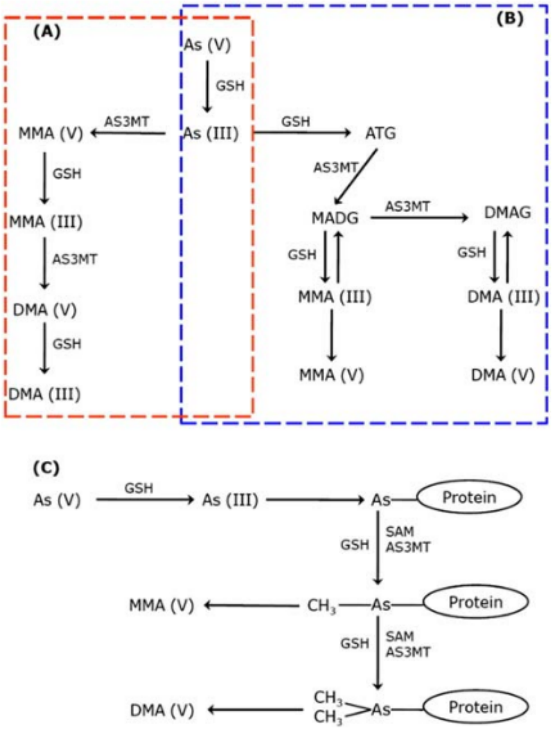

Fig. 1 Common proposed pathways for arsenic metabolism. **a** The classical pathway [30, 31], **b** pathway model proposed by Hayakawa et al. [32], and **c** pathway model proposed by Naranmandura et al. [33] are presented in the red, blue, and black boxes, respectively. GSH, glutathione; AS3MT, As(III) methyltransferase; ATG, arsenic triglutathione; MADG, monomethylarsenic diglutathione; DMAG, dimethylarsinic glutathione; SAM, *S*-adenosyl methionine. This figure is adapted from Bhowmick et al. [29]

(MMA), and dimethylarsinic acid (DMA), or various organoarsenicals depending on environmental conditions. The forms of As(III) and As(V) have been observed to be more dominant in natural waters with more toxic and higher mobility compared to other forms [34]. Moreover, it has been observed that the As(V) species is negatively charged while the predominant As(III) species is neutral in charge in water at pH ranging from 4 to 10 [35]. Redox potential (E_h)-pH diagram is commonly used for the describing the speciation of arsenic at different E_h and pH values as depicted in Fig. 2a [36]. In aqueous systems, protonated oxyanions might be formed from As(III) and As(V) with the pH of the medium that determines the degree of protonation. In oxygenated water systems, arsenious acid (H_3AsO_4) becomes more dominant at low pH (<2) while $H_2AsO_4^-$ and $HAsO_4^{2-}$ are the major species at pH ranging from 2 to 11. In reducing environments, H_3AsO_4 is probably converted to $H_2AsO_3^-$ at low pH while H_3AsO_4 can be in the form of $HAsO_3^{2-}$ at the pH value exceeding 12. For As(III), it can be seen from Fig. 2b that after pH exceeding 6, the distribution species of H_3AsO_3 starts to decrease up to near zero at pH 11 but $H_2AsO_3^-$ becomes more dominant before decreasing after pH 10.5. Moreover, the distribution of the species of As(III) in the form of $HAsO_3^{2-}$ can be observed after pH 10 as shown in Fig. 2b. As a comparison, the distribution of the species of As(V) in the form of H_3AsO_4 and $H_2AsO_4^-$ becomes to zero after pH 4 and pH 8, respectively. Moreover, a complete overview of distribution of the species of As(III) and As(V) at different pH values can be seen in Fig. 2b and c, respectively.

In aerobic conditions, the As(III) is thermodynamically unstable, leading to the formation of a less mobile form of As(V) via oxidation mechanism depending on oxidant type and other redox-sensitive species. The formation rate is relatively slow when the oxygen is the only oxidant, but it can be improved in the presence of redox-sensitive species such as microorganisms, ferric iron (Fe(III)), and manganese oxides (MnO_2). For

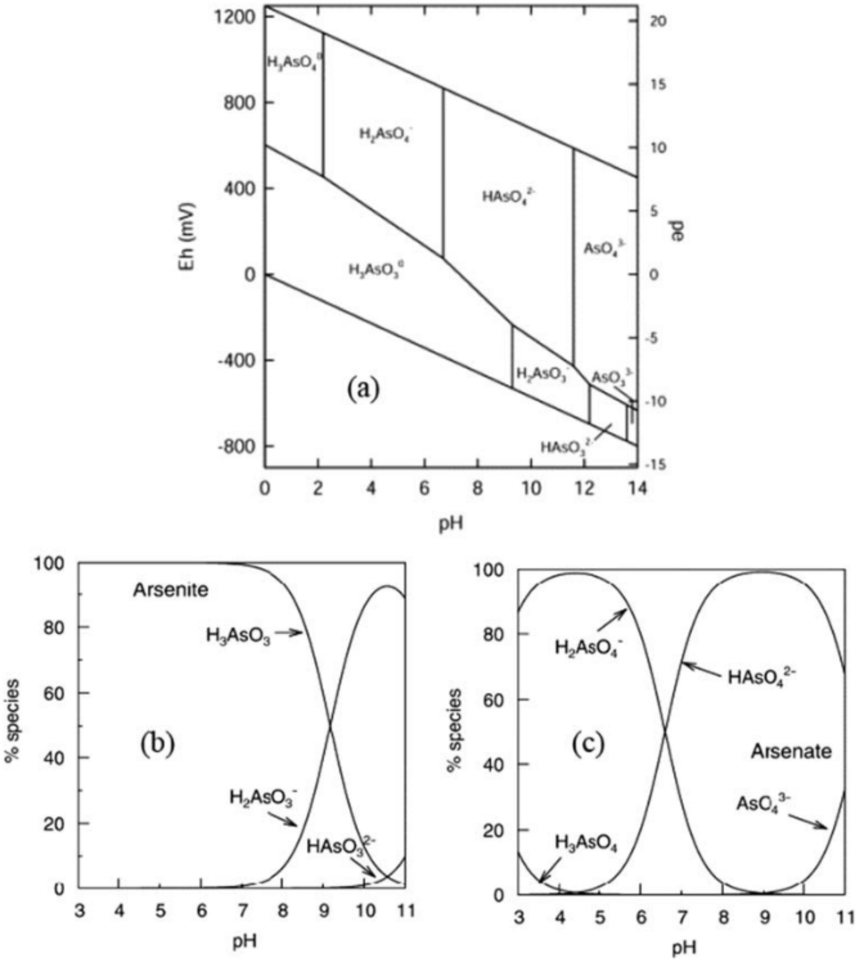

Fig. 2 **a** E_h-pH diagram of arsenic species in water at a temperature of 25 °C. **b** Distribution of the species of As(III) as a function of pH. **c** Distribution of the species of As(V) as a function of pH. The figures are adapted from Smedley and Kinniburgh [36]

instance, the presence of microorganisms such as bacteria can catalyze the oxidation of As(III) and the oxidation reaction can be expressed as [37]

$$H_3AsO_{3(aq)} + \frac{1}{2}O_{2(g)} \xrightarrow{\text{oxidizing bacteria}} H_3AsO_{4(aq)} \tag{1}$$

Moreover, the presence of Fe(III) in aqueous solutions can accelerate the oxidation of As(III) at pH below 7 while MnO_2 can improve the rate of oxidation over a wide range of pH values [38]. The reaction of As(III) with manganese(IV) oxides and manganese(III) oxides can be expressed as [39]

$$H_3AsO_3 + MnO_2 \rightarrow HAsO_4^{2-} + Mn^{2+} + H_2O \tag{2}$$

$$H_3AsO_3 + 2MnOOH + 2H^+ \rightarrow HAsO_4^{2-} + 2Mn^{2+} + 3H_2O \tag{3}$$

The occurrence of nitrate in groundwater possibly acts as the main mechanism for the oxidation of arsenic as this phenomenon has been clarified by the previous work [40]. Figure 3 shows how the presence of nitrate can affect the fate of arsenic in groundwater [41–45]. The data presented in the figure clearly shows that the increase in the nitrate concentration corresponds to the decrease in arsenic concentration. Although water systems are at anoxic condition, the presence of nitrate can promote the formation of arsenic(V) because nitrate can act as a terminal electron acceptor during the process [40]. A field study exhibited how the presence of nitrate can affect the arsenic mobility. A previous study showed that the formation of iron oxides in the groundwater was observed because of the presence of nitrate and it caused the attachment of arsenic on the iron oxides [46]. Interestingly, the study also found that when the nitrate was removed from the water, iron(II) and arsenic(III) can be dominant species as being observed in the pre-experimental conditions. In addition, the addition of nitrate can attenuate arsenic in contaminated groundwater as observed in a study conducted in Bangladesh [47]. The oxidation of As(V) to As(III) to partial denitrification of nitrate to nitrite can be described with the reaction as follows [48]:

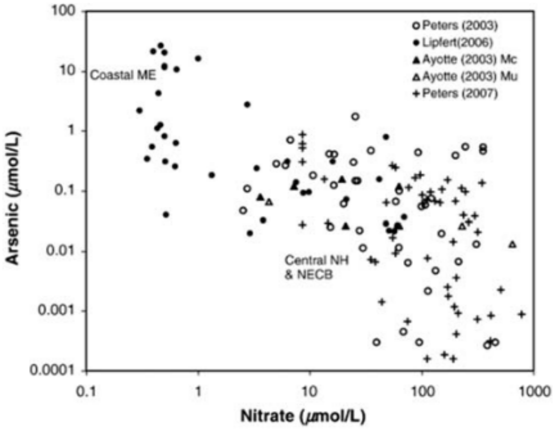

Fig. 3 Plot of arsenic concentrations as a function of nitrate concentrations in the groundwater adapted from Peters [42]. The presented data are as follows: blank circle [45], black circle [43], black triangle [41], blank triangle [41], and cross [44]

$$H_3AsO_3 + NO_3^- \rightarrow 2H^+ + HAsO_4^{2-} + NO_2^- \tag{4}$$

Moreover, the metabolism of As(III) can also be linked to the complete denitrification of nitrate to dinitrogen (N_2) gas with the reaction as follows [48]:

$$5H_3AsO_3 + 2NO_3^- \rightarrow 8H^+ + 5HAsO_4^{2-} + N_2 + H_2O \tag{5}$$

Nitrate contamination in groundwater has become a severe issue worldwide due to the exponential increase of human population and the increase in the demand of water supplies for agricultural activities [49]. Figure 4 illustrates the possible leaching mechanism of nitrate into karst groundwater within the nitrogen cycle [50]. In North America, groundwater serves as the main domestic supply of nearly 80% of the rural population [51]. Inorganic nitrates are most commonly present in the forms of NO_3, NO_2, and NH_4 in soil while those in the forms of plants are usually NO_3 and NH_4. In groundwater supplies, NO_2 and NH_4 are the most commonly present at very low concentrations as they can be swiftly converted to nitrate [52]. The main use of nitrate is in fertilizers for the purpose of enhancement of agricultural productivity simultaneously with other various point and non-point pollutions which have aggravated the negative impacts on groundwater supplies [53]. In Japan, nitrate pollution in groundwater supplies have exceeded the permissible concentration of 10 mg/L due to livestock waste and chemical fertilizers excessively utilized by farmers [54]. Moreover, nitrate pollutants are very soluble and can be passed through the soil structure which then finally gets into the groundwater systems [51].

The export of nitrate into surface water can also easily result in various ecological and environmental issues such as eutrophication and hypoxia which can disrupt the biodiversity of the ecosystems and reduce the oxygen levels of aquatic life. In the context of human health, the long-term presence of nitrate in food and water supplies has the potential to result in birth defects, cell mutation, gastric and esophageal cancer, heart diseases, and methemoglobinemia [55]. It is reported that infectious waterborne disease related to chemical pollutions which include nitrate is on top of the chart for the death of young annually [56]. Hence, human health and the ecological systems are severely affected by the pollution of nitrate in the groundwater. Excessive nitrate utilization should be significantly cut down and banned by the law for a sustainable environment for a better future.

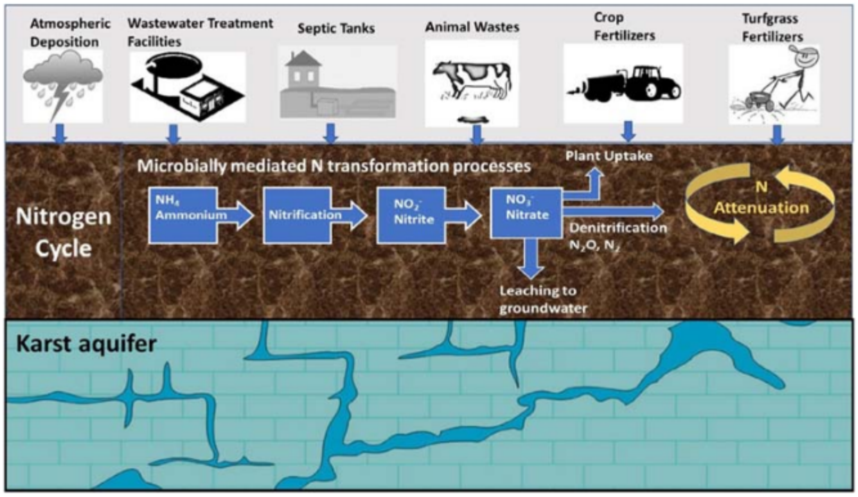

Fig. 4 Possible leaching mechanism of nitrate into karst groundwater within nitrogen cycle [50]

Organochlorine pesticides are among the extremely toxic compounds as they have a high persistence and can be accumulated in human and ecological system. The toxicological effects of organochlorine pesticides were found to cause the etiology of diseases in human which include carcinogenicity and mutagenicity [57]. For example, it has been reported that approximately 3 million severe intoxications and more than 200,000 deaths per year globally were caused by organochlorine pesticide pollution [58]. Reproductive defect, allergies, neurological disorders, and cancer have all been associated with excessive organochlorine exposure [59]. There also have been reports that organochlorine pesticides act as endocrine disrupters which cause neurotoxic damage, low sperm count, asthma, and congenital malformations [60]. However, many developing countries globally still utilize organochlorine pesticides for agriculture and livestock activities and for controlling vector-borne diseases which threaten human health such as malaria and dengue [51]. As a result of the persistent use of organochlorine products, numerous natural means such as water, soil, and air have been polluted including groundwater resources [61]. This associates to the creation of new diseases by pesticides which can lead to a widespread of harm to human health. It has also been reported that the exposure to pesticides has been linked to Parkinson's disease. Several studies have found a significant association between the duration of pesticide exposure and Parkinson's disease with the risk ranging from 2.0 to 3.4 [62] as reported in the case-control study in Taiwan [63] and in Germany [64]. In addition, a study in Canada observed a higher prevalence of Parkinson's disease in rural agricultural regions compared to urban areas [65] and increased mortality due to Parkinson's disease in California because of their high exposure to pesticides [66]. In general, a comprehensive review regarding case studies on Parkinson's disease related to pesticide exposure was reported by Freire and Koifman [62].

Organochlorine pesticides use the water as their main media to spread [67]. This becomes a major concern as nearly 25% of the global population of America, Europe, and Asia uses groundwater mainly supplied by karstic aquifers which are extremely vulnerable to contamination [58]. Karst aquifers are highly vulnerable to contamination as the water in the karst aquifers moves through sinkholes, underground caves, and channels which act as main flow paths of water while moving through the karstic soil [68]. Hence, the karstic soils are capable of acting as the fulcrum for the introduction of organochlorine pesticides into the groundwater system. In Mexico, soil samples collected from the entire Mexican region have displayed that organochlorine pesticides have been utilized and could be potentially the origin of on-site contamination or be conveyed when dehydrated [69]. Thus, the vulnerability of the karst aquifer allows the entrance of organochlorine pesticides into the groundwater system which can cause major health impacts to human health due to the drinking of water contaminated with organochlorine pesticide pollutants.

In Situ Groundwater Pollution Remediations: Advantages and Drawbacks

In situ bioremediation is one of the most endorsed groundwater treatment techniques globally as this method utilizes microorganism to degrade contaminants to less harmful products [70]. In the USA, the utilization of in situ bioremediation in all groundwater treatment methods was approximately 30% for the past several years [71•]. In this procedure, there are two methods that are commonly used, which are biostimulation and bioaugmentation. The biostimulant method generates substrates which are able to provide suitable conditions that can enhance the microbial growth. Another possible implementation of bioremediation is by bioaugmentation where microorganism can be introduced into the groundwater contaminations as shown in Fig. 5a. Furthermore, in situ bioremediation is also very flexible as it can be combined with other remediation methods such as permeable reactive barriers. A study has reported that the integration of polyhydroxybutyrate with zerovalent iron was able to stimulate a very reactive biological reductive dechlorination process [72].

In situ thermal remediation technologies such as electrical resistance, conductive heating, and steam-based heating have also been implemented for the remediation of contaminated groundwater. For instance, removal of chlorinated volatile organic compounds from groundwater was successfully carried out using thermal conductive heating [73]. In addition, pentachlorophenol can also be successfully removed by steam-based heating, which was conducted in a pilot study [74]. In general, the performance of in situ thermal remediation is reviewed by Triplett Kingston et al. [75]. Figure 5b shows an example of in situ thermal remediation for the remediation of contaminated groundwater [76].

Permeable reactive barriers are considered as a passive in situ remediation method as the permeable reactive barriers permit groundwater to move freely in subsurface barrier integrated with reactants for the deterioration and elimination of pollutants [77]. Permeable reactive barriers are considered as an alternative improvement to the traditional pump and treat method as it generates less environmental footprints, helping the sustainability of the environment. Moreover, permeable

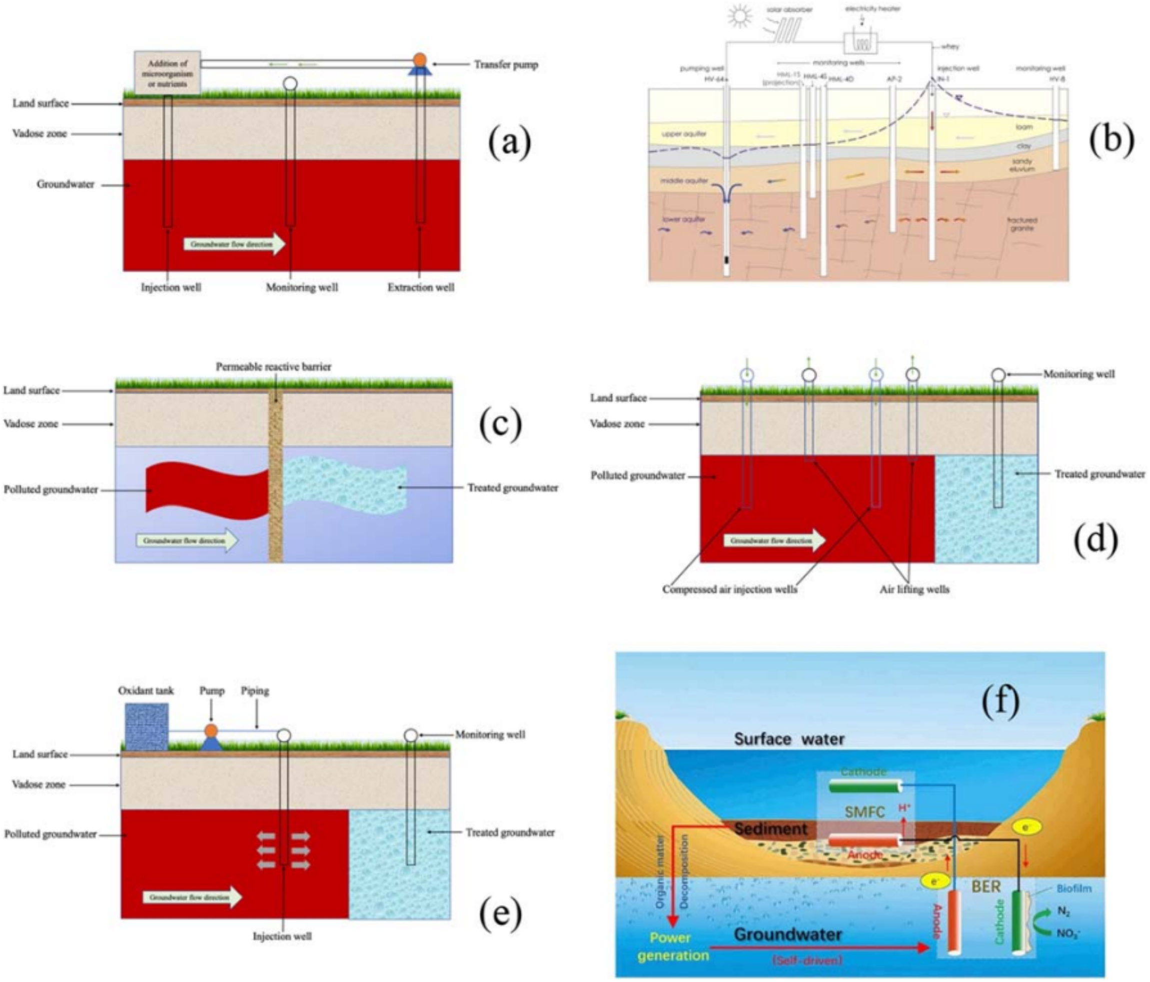

Fig. 5 Schematic configuration of different in situ groundwater pollution remediations. **a** Bioremediation. **b** Thermal [76]. **c** Permeable reactive barriers. **d** Air sparging. **e** Chemical oxidation. **f** Bioelectroremediation [94]

reactive barriers strictly follow the concept of a green and sustainable remediation technology and have been considered as one of the best greenest remediation methods globally. To construct these permeable reactive barriers, a long small trench is required to be dug up followed by introducing containers filled with reactive materials into the subsurface while deliberately placing low-permeability barrier walls which are able to direct the groundwater flow into the containers in order to effectively eliminate the pollutants inside the groundwater system as depicted in Fig. 5c. The reactive metals utilized to react with pollutants are usually zerovalent iron [78]. Other possible reactive media are usually not economical compared to the zerovalent iron, but such new media may be more economical if integrated together with the zerovalent iron as it can enhance the general durability and reaction of the permeable reactive barriers [79]. In general, there are two configuration types of permeable reactive barriers used in the field application, which are the funnel-and-gate design and the continuous gate design. The funnel-and-gate design consists of the funnel designed for converging the plume to the treatment zone before treating at the reactive gate. The configuration of the continuous gate is commonly designed with the placement of the treatment barrier across the entire contaminant path. It has been well known that the use of the continuous gate configuration is more promising because it is easy to construct, is less expensive, and has little effect on the groundwater flow compared to the funnel-and-gate design [80].

In situ air sparging has been implemented for the remediation of contaminated groundwater [81]. In the treatment process, it can be carried out by injecting air in saturated soils as shown in Fig. 5d. It can remove both pollutants in dissolved and adsorbed phases and facilitates the oxygen transfer into the groundwater systems. The treatment can be used for the enhancement of remediation of contaminants by physical as well as by aerobic processes. In some cases, the dissolved groundwater concentrations can be removed during sparging treatment. However, the concentration of pollutants can be back to nearly original levels when the system is turned off, which is commonly known as rebound. It is noted that the performance of this treatment system is highly dependent on the contact between the injected air and the contaminated groundwater. The performance of these systems for the groundwater remediation was comprehensively reviewed by Bass et al. [82].

The implementation of in situ chemical oxidation (see Fig. 5e) is normally seen in the petroleum industries for the treatment of petroleum hydrocarbon, and it is considered as one of the most implemented remediation methods [83]. With reference to the US superfund program, the implementation of in situ chemical remediation has also been increasing, which is approximately 20% of all groundwater treatment methods [71•]. The implementation of in situ chemical oxidants includes hydrogen peroxide and permanganate or alternatively by ozone or persulfate [84]. Recent in situ chemical oxidation technologies face issues related with the non-selective consumption of oxidants by soil substances. A study has reported that only an estimated 20% of permanganate utilized only reacted in oxidation reaction with the contaminant tetrachloroethane [85]. Thus, to ensure tetrachloroethane is efficiently removed and eliminated, permanganate and tetrachloroethane are required to be at a ratio of 82:1.

In situ bioelectroremediation or commonly known as microbial electrochemical technology (MET) has been recently proposed for the remediation of groundwater. This treatment combines the use of microbiology and electrochemistry and is found to be a reliable and effective procedure for remediation of contaminated groundwater. The electrode is utilized as electron acceptor or electron donor. This can be alternative to oxygen/nitrate or organic matter/hydrogen in the conventional chemical treatment, respectively. In this method, the remediation is initiated by injection of electrodes into groundwater system to stimulate the native microorganisms [86]. This treatment technology has been applied for the removal of aromatic hydrocarbons or dissolved metals [87–89] or nitrates, metals, and chlorinated hydrocarbons [90–92]. The performance of bioelectroremediation for the groundwater remediation was reviewed by Cecconet et al. [93]. Figure 5f illustrates the configuration of this system for the remediation of contaminated groundwater [94]. This system has currently been successfully applied for the remediation of groundwater containing toluene and ethylbenzene [95]. The study found that the proposed method can remove the pollutions with degradation rates by 31.3 ± 1.5 mg/L/day and 3.3 ± 0.1 mg/L/day for toluene and ethylbenzene, respectively. For confirming a clear mechanism and identifying possible metabolic intermediates, gas chromatography-mass spectrometry (GC-MS) analysis was conducted and found that the presence of benzylsuccinate, which is a typical product of anaerobic toluene activation via the fumarate addition pathway, was detected after the treatment. In addition, 1-phenylethyl-succinate was also detected after the remediation, which is the metabolic intermediate resulting from anaerobic ethylbenzene activation. It is noted from the study that the identified metabolites (benzylsuccinate and 1-phenylethyl-succinate) are due to the electrogenic activation and the methanogenic activation of toluene and ethylbenzene [95]. A comprehensive overview, advantages, and drawbacks of the selected in situ remediations are listed in Table 1 [14, 73, 80, 93, 96–103].

Challenges of Groundwater Pollution Remediations and Their Potential Solutions

Common challenges of in situ remediation such as back diffusion, tailing, rebound after stopping the treatment, and longevity are illustrated in Fig. 6 [71•]. The presence of pollutants

Table 1 Advantages and drawbacks of existing in situ remediation technologies for contaminated groundwater

Remediation method	Advantage or drawback		Reference
HE	Advantage	Increases performance of the technology used in post treatment	[73]
	Drawback	Necessarily followed by another remediation technology	
		High energy consumption	
AS	Advantage	Simple, rapid, and economical	[103]
	Drawback	Not applicable to non-volatile contaminants	
		Not suited for confined aquifers	
PRB	Advantage	50% cheaper than the pump and treat technique, as no routine maintenance and surface buildings	[98]
		Intercepts groundwater flow, long-lasting, variety of setups and solutions	[80]
	Drawback	When dissolved contaminants come into contact with the PRB, numerous reactions can occur that gradually reduce the removal efficiency, porosity, permeability, and longevity	[97, 100]
		Fouling can also reduce PRBs' porosity and hence hydraulic conductivity	[99]
		Strongly dependent on the site characteristics	[80]
		Necessary addition of chemicals/nutrients	
BIG	Advantage	Pre-adapted bacteria are able to remove target contaminants and mimic natural process	[96]
	Drawback	Long remediation time	
		Not suitable for large scale	
		Need the addition of nutrients and oxygen	
BIS	Advantage	Use of naturally present bacteria	[101]
	Drawback	Not suited for highly polluted sites	
		Addition of nutrients and oxygen supply may be costly in the long period	
CO	Advantage	Highly effective	[93]
		Involvement of toxic chemicals	[14]
	Drawback	High cost	[93]
BIE	Advantage	No addition of chemicals, electrodes acting as electron donor/acceptor, use of indigenous microbial consortia; low energy demand can be supplied by renewable sources	[102]
	Drawback	Still at laboratory or pilot scale	

HE heating, *AS* air sparging, *PRB* permeable reactive barriers, *BIG* bioaugmentation, *BIS* biostimulation, *CO* chemical oxidation, *BIE* bioelectroremediation

in low-permeability zones such as silt and clay has currently been recognized as a major problem in the remediation of contaminated groundwater. These zones polluted with contaminants can potentially act as long-term contaminant reservoirs, spreading the contaminants into the groundwater system through the back diffusion process [104]. Hence, the current trend focuses how the remediation technology can be capable to handle the remediation not only in high-permeability zone but also in low-permeability zone. Although some remediation methods have been applied, this challenge is an unresolved challenge. For instance, the implementation of an in situ chemical oxidation (ISCO) by using permanganate cannot solve this problem since the permanganate only penetrated a maximum of 1.3 cm into the edges of the silt layers, suggesting that remediation of the silt itself cannot be effectively achieved [105].

Hence, bioelectroremediation has been recommended and become an alternative approach to solve the challenge [106, 107]. In this method, the implementation of electrokinetics can be a promising way for increasing the effectiveness of remediant such as nano-scale zerovalent iron, permanganate, or persulfate delivered into low-permeability zones. In general, these studies confirmed that the penetration of permanganate can be improved with the combination of electrokinetics. Cang et al. [106] found that the bioelectroremediation (combination between permanganate and electrokinetics) successfully degraded 52% of pyrene after 336 h. Another study also found that 91% of phenol was removed during 120 h using similar method [107].

Rebound is a phenomenon whereby the pollutant concentrations are reduced significantly during the remedial activities, but the contaminants tend to increase after the remediation system is stopped. Rebound commonly occurs due to the mass transfer and insubstantial transport [108]. In the USA, ISCO project found that the mass emission rate of tetrachloroethane was noticed to have doubled within a year after the permanganate injections were ceased [109]. An alternative method to solve the problem is by implementing ISCO by surfactant combined with RemMetrik® as well as Wavefront's Sidewinder™ tool [110]. The study successfully

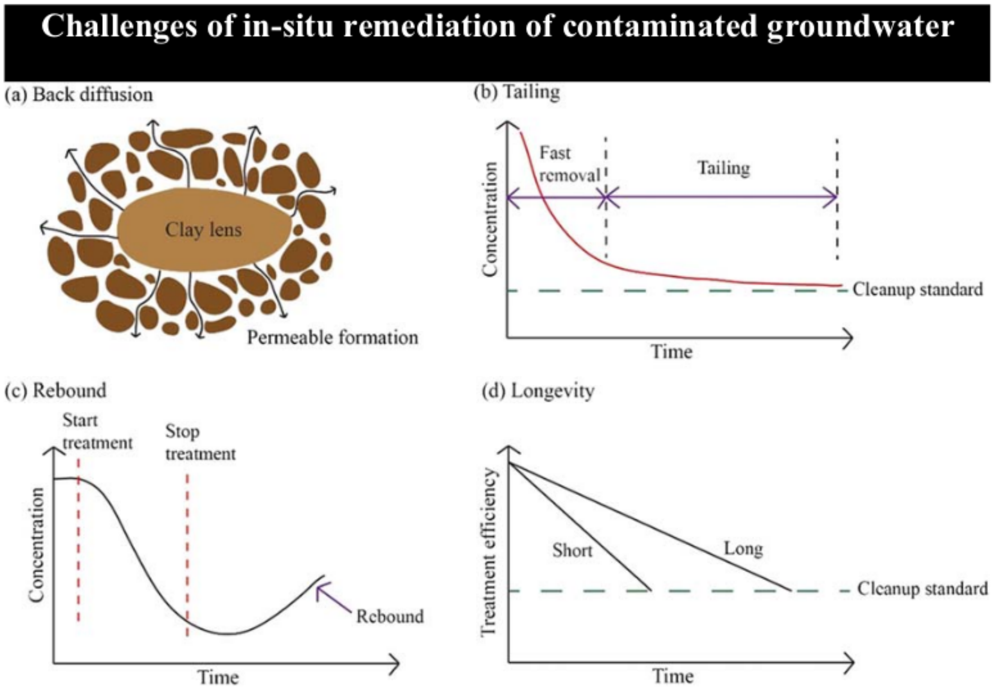

Fig. 6 Common challenges of in situ remediation of contaminated groundwater. **a** Back diffusion. **b** Tailing. **c** Rebound after stopping the treatment. **d** Longevity. This figure is modified from O'Connor et al. [71•]

achieved the reductions in post treatment by 49% and 92% for toluene and xylenes, respectively.

It has been well known that the remediation of contaminated groundwater can be significantly achieved in the initial phase of operation, but it tends to be a slow reduction after a continuous long-term operation. This phenomenon is commonly known as tailing. Concentration tailing is well known as the desorption rate limitation, subsurface diverseness, and dissolution rate limitation of non-miscible liquid [111]. A study has reported that the tetrachloroethane pathway was directed by the non-miscible liquid dissolution during the initial phase of remedial treatment, but once the non-miscible liquid was removed, the tetrachloroethane pathway was directed by rate-limiting desorption which leads to the long-term low-concentration tailing [112]. In addition, a simulated study has shown that concentration of tailing may have occurred due to low conductivity in aquifers [113].

Permeable reactive barrier is developed to repeatedly treat groundwater pollutants for a given period of time. Another important parameter is the longevity of the barrier, which is described as the time for the system to continuously treat contaminants at designed levels. The most customary type of permeable reactive barrier material is zerovalent iron which has drawbacks regarding the long-term hydraulic properties. In addition, removal inefficiency has also been reported and this is possibly due to the deactivation, corrosion, and clogging of the barrier pores [114]. For instance, numerous dissolved components within the groundwater such as magnesium, oxygen, calcium, arsenic, cadmium, and sulfates can react with the zerovalent iron which affects its reactivity [115]. With the introduction of oxidation or precipitation of other minerals, the clogging of pores can occur which helps in the enhancement of the longevity of the reactive media [80]. Intensive works have been carried out to find high-longevity reactive media. For instance, zerovalent iron paired with iron sulfide was proposed as alternative reactive media due to the fact that iron sulfide is thermodynamically more stable which is much unlikely to suffer a huge reduction in permeability [116]. Alternatively, zerovalent iron can be mixed with other reactive barriers such as zeolite and activated carbon [117]. The study found that the optimum mixing was 50% for zerovalent iron, 10% for zeolite, and 40% for activated carbon for a practicable longevity of ≥ 10 years. Mixing between zerovalent iron and trichlorethylene was found to have capability for the degradation of trichloroethylene, which was found to be three times faster than zerovalent iron alone [14].

Integrated Management: Challenges in Managing Groundwater

Integrated management coordinates the groundwater management and related groundwater resources [18]. It has taken into account the interaction of the non-groundwater policy with the purpose of achieving the three pillars of sustainability outcomes through balancing the economics, social welfare, and protecting the environment over space and time [118]. Integrated assessment is a meta-discipline for the integrated groundwater management process [119]. As to apply the integrated groundwater management, a scientific meta-discipline is required to solve the management challenges with sufficient knowledge and access the knowledge towards the social learning for a better process in decision making [118]. Through the integrated assessment, the problems in the public policy which involves the long-term environmental management that affects the groundwater management can be found [18]. In addition, integrated assessment has also been developed in the management for climate change, acid rain, water and air quality management, public health, aquatic management, and land degradation. Integrated groundwater management and integrated assessment are important in achieving efficiency and useful outputs for a sustainable development [18]. There are several key dimensions of integrated groundwater management which include the concern issues, governance arrangement for management, and stakeholders. These dimensions also become the challenges for groundwater management [120].

Many issues in managing the groundwater are interconnected and cannot be solved in isolation [18]. For example, the traditional gravity irrigation systems would not provide maximum recharge of groundwater for further usage [121]. This irrigation system is not modernized by the groundwater managing department such as in India. The groundwater resources are only used once by the villagers in India and not reused or recycled for further uses such as agriculture which leads to the overexploitation of groundwater resource [122]. The policy interventions also cause the concern issues as the policy designed initially to solve the groundwater management issues has disturbed with other groundwater activity policy [121].

The continuous enforcement in pumping restrictions for limiting use of groundwater resources has led to the drastic changes in crop production and the increasing competitiveness between local agricultural industries [123]. Thus, isolating and addressing the groundwater problems can inadvertently create more or aggravate other problems in managing the groundwater systems. In order to avoid the adversely offsetting actions or improve the issues of concern, a joint assessment and the treatment for the issues among the policy segments, such as climate change adaptation, water supply, industry and urban pollution management, environment, agricultural activities, and land development and planning sector, are very important to overcome the poor groundwater management [18]. For instance, the government of Malaysia is currently developing the Malaysian Climate Change Adaptation Index for the evaluation of all states in Malaysia in dealing with the possible climate change impact based on vulnerability and readiness indicators covering various sectors such as water resources, water industry, environment, governance, social, and economic. The Ministry of Energy and Natural Resources has been responsible for the development and assessment. The target of this index is the representation of a scatter plot of readiness against vulnerability for comparing all states and tracking their progress through time.

A complete treatment for groundwater management that related to the issues is necessary to ensure conflicts from stakeholders are included and considered [18]. A clearly articulating statement has been considered in the essence of integrated groundwater management, and the making of trade-offs has been included to limit the adverse effects and balance the values and needs [120]. Integrated groundwater management process can involve the selection of suitable environment, economics, or social as assessment criteria. Integrated assessment can be used in this process to model and assess the performance of groundwater system under different situations [18].

The dimension of governance for integration is widespread around the world, but it is often the primary problem in blocking the effectiveness of integrated groundwater management [18]. Groundwater governance controls and protects the utilization of groundwater resources and groundwater or aquifer systems. Groundwater governance is approved by the legal and regulatory framework. Inside the governance, the knowledge and awareness about the sustainability challenges, policies, and beneficial establishments and inducement structures aligned are being shared with the societies' goals. Various perspectives can be used to examine the groundwater governance such as the institutional structure and the participants that involved or accountable in the process.

There are five types of instruments in the governance policy which are the command and control instruments, economic instruments, communication and diffusion instruments, infrastructure instruments, and collaborative instruments [124]. These instruments should be included and developed in the decision-making process to achieve a sustainable groundwater management which delivers appropriable environmental, economic, and social outcomes [18, 124]. These instruments also represent a robust policy under the potential changes of environment and human settings such as the climate change and increase in population. Integrated groundwater management should provide a process that can evaluate their effectiveness under various conditions through identification of decisions and instruments [18]. Groundwater governance is a complex process, and the effectiveness often interrupts with some challenges that related to the implementing policies among the allocation of groundwater. For instance, it was found that the groundwater governance provided some challenges towards the groundwater management in Bangladesh [125]. The groundwater governance policy in Bangladesh is currently improving the outcomes of the industries by excessive use of groundwater resources without permission from policy and the deduction of taxes [125, 126]. These actions have posed threat to the groundwater management for sustainable groundwater consumption.

Moreover, the government of Bangladesh has provided beneficial finance systems to the local farmers in order to increase the rate of agricultural productivity [125]. The irrigation hours then increase rapidly due to the lower cost of electricity, especially in the rural areas which causes the overexploitation of groundwater resources as irrigation systems are highly reliant on the use of groundwater resources [125]. The economy in Bangladesh has been growing constantly in recent years which result in significant use of groundwater [127]. Numerous policies are being introduced to resetting the water allocation and separation [127]. However, the governance policy systems have interrupted the whole groundwater management process and cause more groundwater contamination [125]. Thus, groundwater governance provides advantage to the society but also becomes a challenge in managing the groundwater systems.

Stakeholders such as local public, national government, groundwater users, and the water industries are considered as individuals or groups that elaborated or interested in managing the problem. Engagement process of stakeholders has often been avoided by the scientists in groundwater management. However, the process of stakeholder engagement is an important statement for the efficiency of integrated groundwater management [128]. Through the stakeholder engagement process, the conflicts among the decision makers and other stakeholders can be reduced. This engagement process also considered as a valuable process as the process is mutually educational between the researchers or scientists with other stakeholders [128]. Stakeholder's engagement also helps to build or develop a wider understanding about the groundwater demands and publicizes scientific information for a better groundwater management. In Australia, it was found that each different jurisdiction has their own statutory consultation organizations in groundwater management planning process [118]. However, the consultation about the planning process often appears more illustrative than reality. In the decision making, stakeholders are highly rated with the intercorporate in participative modeling, assessment, and planning procedures [118].

Integrated groundwater management involves the human setting such as cultural, social, economic, and political factors. The population which is also considered as the human setting has also become a challenge in managing the groundwater resources [129]. As the population increases, the demand for groundwater resources increases. For instance, the population in China is increasing rapidly which causes the formation of overexploitation of groundwater resources and the process to manage the groundwater systems or groundwater resources become more difficult [130]. It was found that due to the growing population in China, the amount of groundwater resources usage has risen for the anthropogenic activities such as construction and agricultural activities, which caused 16% of the country's soil being polluted and 83% of the soil contaminated, and also formation of the groundwater contamination occurred [130].

Solutions for Improving Integrated Management

Various types of methods can be used to improve and support the development of policy in integrated groundwater management. Development of conceptual models against the stakeholder's sector has been used as a framework to solve the related problems, define outcomes, and manage the complexity of groundwater systems [128]. The initial step to solve the management challenges is to plot wide-system boundaries in the managing process, to encompass the interacting impacts. Integrated models are commonly stated as a primary tool to examine and articulate the conceptuality of the groundwater management. Integrated models are represented as a potential framework to minimize the interventions inside the governance policy, uncontrolled human setting or natural setting such as climate change, and the unpredictable outputs or uncertainties [128, 131].

Integrated models capture the trade-offs and influences of alternative activities or actions towards the unsustainable usage of groundwater resources. When the integrated models are constructed properly, the system feedbacks of the groundwater can be explored, and the linkages between the single frameworks can be detected with the effective integrated models [128, 132]. Integrated models are considered as a useful model for the integrated groundwater management process as integrated groundwater management consists of a broad range of human setting, positive or negative opinions, and spatial-temporal scales. Thus, integrated models are often applied in the integrated groundwater management process for various groundwater system components [18, 133]. For instance, an integrated model was proposed by exploring the ecological impacts and socioeconomics among the rural farmers about the allocations of water reduction and adaptation options with the reduction [134]. The model was developed as a groundwater surface model with various opinions which involved the ecological expert opinion, policy rule models, social Bayesian networks, and crop meta-models [134].

A modeling framework and process is needed for the integrated assessment which included the requirement of integrated perspectives from various stakeholder groups with the different disciplines, in order to present an adaptive and encouragement in the participatory procedures [133, 135]. A flow of information can be provided to the environmental policy makers about the knowledge of stakeholders towards the groundwater management systems and preferences. This information shows a support to the conceptualization, construction, and the use of integrated model from the stakeholders to the groundwater management policy, which can improve the understanding between the stakeholders and the policy makers in order to achieve a sustainable groundwater management [135]. This is because both the information from the policies and stakeholders can be shared among them. Alternatively, scientists can gain the understanding of the information from the modeling process and thus provide a good feedback and interactions with the stakeholders [18].

Case Study on the Exploration of Groundwater in Malaysia

Due to increasing demand and polluted surface water resources, the government of Malaysia has promoted the exploration of groundwater as an alternative water resource. It was estimated that the groundwater storage in Malaysia is about 5000 billion m^3 with the annual recharge of 64 billion m^3 [136]. The highest rate of groundwater consumption in Malaysia is for the state of Kelantan, which consumes about 160 million liters per day (MLD) or 40% of the total state water usage [137]. In general, the exploration of water resources has been regulated by the government particularly for domestic and non-domestic uses. In the current context, the exploration of groundwater is regulated under the Food Act 1987 regarding contaminations towards the water quality. At the state level, Kedah and Selangor have regulations to control groundwater abstraction and other particular matters such as licensing, penalties, monitoring, and determination of zoning critical areas under Kedah Water Resources Enactment 2008 and Selangor Waters Management Authority Enactment 1999, respectively. Under these regulations, only these states have full authority and power to take any action according to the enactments of any offenses towards groundwater abstraction.

Studies on the evaluation of groundwater contaminants have also been established including the investigation of organic and inorganic contaminants. For inorganic contaminant, the presence of several heavy metal pollutants such as lead (Pb), cadmium (Cd), selenium (Se), aluminum (Al), manganese (Mn), copper (Cu), zinc (Zn), iron (Fe), arsenic (As), nickel (Ni), chromium (Cr), and silver (Ag) has been detected as observed at the groundwater of Lorong Serai 4, Hulu Langat, west coast of Peninsular Malaysia [138]. Among the detected heavy metals, the concentration of Fe, Mn, and As exceeded the permissible limits regulated by the World Health Organization (WHO) and Malaysian Ministry of Health (MMOH) for trace metals in drinking water. The study then proposed magnetite coated with graphene oxide (Fe_3O_4-GO) nanoparticles (NPs) as the adsorbent to remove the pollutants and found to have capability with the removal efficiencies by 99.1%, 39.3%, and 82.9% for Fe, Mn, and As, respectively. Alternative study by using metakaolin as the adsorbent found that the proposed adsorption method can remove Mn by 33.2% within 120 min from the groundwater sample collected from tube well located in the Universiti Sains Malaysia (USM) Engineering [139].

For organic contaminants, their detection in groundwater samples has also been observed. For instance, a study by National Hydraulic Research Institute of Malaysia (NAHRIM) observed the presence of pesticide contamination in groundwater samples collected from North Kelantan at the selected agricultural areas cultivated with paddy and tobacco [140]. The study found that the endosulfane was detected in the groundwater samples and was most widespread in the paddy area compared to tobacco. It was hypothesized that the presence of pesticide in the groundwater samples was because of the extensive use of pesticide for pest control. Alternative study also observed the presence of volatile organic compounds in groundwater collected from various sources in the Peninsular Malaysia [141]. The study proposed constructed wetlands (CWs) with subsurface flow system in combination with *Typha angustifolia* for phytoremediation of volatile organic compounds which was found to be a successful method indicated by the reduction in the water parameters measured.

In Malaysia, Air Kelantan Sdn. Bhd. (AKSB) becomes the only drinking water treatment company that uses groundwater as a raw water source [142]. In general, the remediation of groundwater in Malaysia is carried out using ex situ method. One of the established methods applied in real application is the Nature Groundwater Eco-Treatment (N-GET) installed at Research Centre for Soft Soil Malaysia (RECESS), and the treated water is used for daily activities [142]. The method consists of four treatment tanks, which are aeration, sedimentation, suction, and distribution tanks. In the remediation process, the groundwater collected from tube wells was pumped and channeled into the aeration tank for the removal of carbon dioxide before entering the sedimentation tank for settling sediments for 48 h. The treated groundwater was then filtered using filtration system consisting of gravel sand, ceramic media, and activated carbon for the removal of pollutions before storing into the distribution tank for daily usage.

To improve the groundwater management, the government has promoted several initiatives. For instance, the Department of Mineral and Geoscience (JMG) has strengthened the management via increasing research and providing monitoring services on groundwater quality. In addition, JMG has also provided information on safe rate of extraction to state governments as guidelines in issuing groundwater extraction licenses at the coastal areas to avoid saltwater intrusion.

Moreover, JMG has planned for the development of 4D hydrogeological mapping for selected states such as Kedah, Johor, and Perak under the Twelfth Malaysia Plan, 2021–2025. Currently, participative modeling has been promoted to address challenges in water resources management and this needs the importance of involving stakeholders in a modeling process [143]. In the context of Malaysia, participative modeling is recommended to be carried out for groundwater management since such study is hard to find in the literature. As an alternative, participatory modeling of surface water and groundwater to support strategic planning in the Ganga basin in India can be used as a reference [144]. Moreover, a comprehensive discussion on participatory modeling for water resources management was provided by Basco-Carrera et al. [145].

Future Outlook and Recommendation

The overexploitation of groundwater resources due to the anthropogenic activities such as agricultural activity and industrial activity has led to the formation of groundwater contamination and thus threatened the management of groundwater. The challenges in managing the groundwater system with integrated process have also been introduced which included the issues of concerns on the groundwater sustainability, the groundwater governance policy, stakeholders involved in the usage of groundwater resources and the groundwater systems, and human setting that affect the groundwater management. These challenges can be minimized or solved by developing an integrated modeling tool. However, it was found that there is still a challenge in using the integrated modeling tool to achieve a sustainable integrated groundwater management, which is the effectiveness of the communication between the tools.

If the communication between the tools can be improved, the modeling can facilitate the integrated groundwater management through improving and articulating the understanding between the stakeholders and environmental policy. The education between the scientists, decision makers, and other stakeholders can also be improved. To develop successful integrated modeling tools for a sustainable management, a conjunctive water management can be developed [146••]. Conjunctive water management is the combination of managing both surface water and groundwater resources [133, 146••]. The conjunctive management can be used to determine or examine both surface water and groundwater problems, which can also achieve the public policy and groundwater management [133]. Conjunctive water management is also able to manage greater stability and security of water supplies. The variation of the climate change can also be adapted with this type of water management, thus reducing the degradation or depletion of both surface water and groundwater resources [146••]. Therefore, conjunctive water management can be considered as a better recommendation in achieving a sustainable groundwater management in developing countries.

Conclusion

This paper highlighted the current status of groundwater usage from the point of view of pollutant control and integrated management. Sustainable efforts were suggested to minimize the arsenic content from all the possible sources before entering the groundwater system and to minimize the excessive nitrate and pesticides uses in order to be in accordance with sustainable environment goal. The widespread use of in situ remediation by biological method must be encouraged since it has the capability to reduce cost and the use of toxic chemicals. Conjunctive water management has been recommended to enhance security of groundwater supply and environmental sustainability.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

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