METHODS FOR NITRATE REMOVAL FROM GROUND WATER

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Abstract—

Groundwater is one of the most important sources of drinking water in rural area of India, and in urban area maximum of domestic water is procured from ground water. Therefore India is largest extractor of ground water in the world. When ground water is used for drinking purpose, contamination in groundwater is not looked after seriously. Though groundwater at shallow depth is considered as most suitable for domestic use, but due to continuous extraction of groundwater in last decade in uncontrolled manner and groundwater level gone much deeper in aquifer. The quality of groundwater in deeper aquifer varies from place to place and contamination is becoming more concentrated. As per Central Groundwater Board survey, in 387 districts in various states has contaminated groundwater containing arsenic, fluoride, nitrate and iron. In Maharashtra most of the districts exceeds in Nitrate contamination above permissible limit of 45 mg/l. Because of this government has banned the villages to consume groundwater from such contaminated hand pumps & tube wells. In India for increasing agricultural production huge amount of fertilizers up to 14% of global total is consumed, therefore Nitrate contamination in groundwater is increasing day by day. Concentration of nitrate above 50 mg/l in drinking water generate various health implications such as infant mortality, spontaneous abortion, birth defects, abdominal pain, diarrhea, vomiting, changes in immune system and methemoglobinemia. Because of various sources of nitrate contamination occurring in groundwater Nitrate removal is one of the emerging challenge. There are various methods of removal of Nitrate from wastewater as well as from drinking water. Distillation, reverse osmosis, ion exchange, electrodialysis, blending, chemical denitrification are the in-home methods for removal of nitrate from drinking water. With the advancement in water treatment technologies, adsorption using activated carbon, carbon nanotubes, biological nitrogen and phosphorus removal, membrane techniques are also used now days in many places. Catalytic denitrification, electrocatalytic reduction for denitrification, denitrification using membrane bioreactor, combined ion exchange & membrane bioreactor for denitrification, denitrification using nanofiltration are some of immersing technologies. Reverse osmosis, ion exchange, distillation are the method of nitrate removal requiring high initial cost as well as maintenance cost. On the other hand biological denitrification is promising technology widely used for nitrate removal from municipal wastewater. Despite the development in these technologies, a need remains for a low cost, low maintenance, efficient method to treat nitrate contaminated groundwater.

Keywords—Groundwater Contamination, Nitrate, Nitrification, Denitrification, Nitrate Removal

I. INTRODUCTION

Groundwater is one of the most important water sources in India accounting for 63% of all irrigation water and over 80% of the rural and urban domestic water supplies. In fact, the United Nations Educational, Scientific and Cultural Organization World Water Development Report state that India is the largest extractor of groundwater in the world. Fifty-four percent of India’s groundwater wells have declined over the past seven years, and 21 major cities are expected to run out of groundwater by 2020. Thus, India faces a dual challenge: to regulate the growing demand for groundwater while replenishing its sources. On the other hand whatever ground water is used for drinking water, it is contaminated with various contaminants. And thus ground water contamination is other bigger challenge for India.

Indian Sub-Continent is endowed with diverse geological formations from oldest Achaean to Recent alluviums and characterized by varying climatic conditions in different parts of the country. The natural chemical content of ground water is influenced by depth of the soils and sub-surface geological formations through which ground water remains in contact. In general, greater part of the country, ground water is of good quality and suitable for drinking, agricultural or industrial purposes. Ground water in shallow aquifiers is generally suitable for use for different purposes and is mainly of Calcium Bicarbonate and Mixed type. However, other types of water are also available including Sodium Chloride water. The quality in deeper aquifers also varies from place to place.
I. Health Effects of Ground Water Nitrate Pollution

High concentration of Nitrate in water beyond the permissible limit of 45 mg/l causes health problems. Nitrate exposure can lead to several health problems such as spontaneous abortion, increased infant mortality, birth defects, abdominal pain, diarrhea, vomiting, diabetes, hypertension, respiratory tract infections, changes in the immune system, and methemoglobinemia. To limit the risk to human health from nitrate in drinking water, World Health Organization and the European Community have set the Maximum Acceptable Concentration and Maximum Contaminant Level is set to be 45 mg/L. There are many health effects of nitrate pollution and therefore exploring methods for nitrate removal from ground water is required. Globally, among the selected chemical threats to groundwater, nitrate (NO3-) is listed as second most common pollutant of groundwater next to pesticides. Nitrate concentrations are increasing rapidly at high rates leading to concentration of above 45 mg/L and generating health hazards especially for babies and pregnant women. The most threatening of all the health impact is ‘Methemoglobinemia’ also known as ‘Blue Baby Syndrome’ which is caused when water with high nitrate concentrations ingested in infants especially in “bottle fed neonates”.

Initial Literature survey revealed that nitrate also causes diabetes and is a precursor of carcinogens. Several studies suggested that, at levels between 100-200 mg/l, nitrate-nitrogen in water causes decrease in appetite in livestock and asks for better management practices to prevent livestock loss. Ingestion of nitrate contaminated plants or water can cause acute poisoning in livestock within 30 min to 4 hours. The World Health Organization recommended limits are 50 mg/l as nitrate ion (or 11 mg/l as nitrate-nitrogen) to protect against methemoglobinemia in bottle-fed infants (short-term exposure). Whereas, the acceptable limit given by Bureau of Indian Standard (BIS) is 45 mg/l as nitrate. The guideline value 50 mg/l as nitrate is based on epidemiological evidence for methemoglobinemia in infants, which results from short-term exposure and is protective for bottle-fed infants and, consequently, other population groups.

Several health problems may be caused by excess nitrate in water sources. Normally, nitrate is eliminated through the kidneys before converting to nitrite, but it is reported that high intake by pregnant women can cause spontaneous abortion and birth defects such as neural tube defect. In adults with reduced stomach acidity or deficient in the methemoglobin reductase enzyme, high amounts of nitrate may cause abdominal pain, diarrhea, vomiting, diabetes, hypertension, respiratory tract infections, and changes in the immune system. Nitrate is converted to nitrite through microbial reduction. The reaction between nitrite and secondary or tertiary amine in acidic mediums such as the human stomach can result in the formation of N-nitroso compounds, which are known to be carcinogenic, teratogenic, and mutagenic. N-nitroso compounds might cause cancers such as stomach and bladder cancer. However, studies that investigated relationships between drinking water nitrate contamination and cancer risks have resulted in contradictory conclusions. Most important health concern associated with nitrate is that it causes methemoglobinemia, a potentially fatal disorder, in infants under six month of age. Methemoglobin (MetHb) is a form of hemoglobin (Hb) that cannot bind oxygen. Nitrite transforms Hb to MetHb by oxidizing the ferrous iron in hemoglobin to the ferric form. However, based on the limited data, it is not possible to specify an exact level as a safe nitrate intake level for all infants. On the other hand, it is also reported that many cases of methemoglobinemia in infants might be caused by overproduction of nitric oxide due to gastrointestinal infection and inflammation and not by consuming drinking water nitrate.

II. METHODS FOR NITRATE REMOVAL

Nitrate easily dissolves in water, and it is very difficult to remove. If water contains more than 45 ppm of nitrate-nitrogen, the viable options for reducing health risks are drinking water replacement or in-home treatment. Unfortunately, simple household treatment methods for other contaminants such as boiling, filtration, disinfection, and water softening are not effective for nitrate removal. Boiling will actually increase the nitrate concentration of the remaining water. The use of bottled water for cooking and drinking is a relatively inexpensive method for reducing the health risks associated with nitrate intake. The three in-home water...
treatment methods that can effectively remove nitrate are distillation, reverse osmosis and ion exchange. Treatment of drinking water to remove nitrate is expensive. Factors to consider when purchasing a treatment system are the initial purchase price, the cost of regular maintenance, electricity rates and the quantity of drinking and cooking water required.\[9\].

i) **Distillation**

Contaminated water is heated to form steam. Nitrate, as well as other inorganic compounds and non volatile organic molecules, do not evaporate with the water and are left in the boiling tank. The steam is then cooled and condenses in a separate tank to form clean drinking water. Large distillers can distill approximately 1.5 liter water per hour. Small units produce roughly 1.25 liter of water per hour. Operating cost of such distillation unit depends on the particular system and the cost of electricity. Initial costs range from 17500/- for small unit to over 105000/- for the largest in-home unit. Electricity consumption makes operating costs higher than the other treatment methods. Distillers are relatively simple systems but require periodic maintenance to remove scale from the boiling tank. Maintenance intervals vary with the quantity of contaminants in the water and the amount of clean water required.

ii) **Reverse Osmosis**

Reverse osmosis involves the movement of water under pressure through a membrane. The membrane has macroscopic openings that allow only water molecules to pass through, leaving behind nitrate and other inorganic chemicals like calcium and magnesium. [10] Unfortunately, reverse osmosis also removes beneficial chemicals, such as fluoride. The initial cost of reverse osmosis unit ranges from 21000/- to 91000/- depending upon the amount of water needed. Maintenance costs include periodic replacement of the reverse osmosis membrane. In the case of reverse osmosis water passes through a semi-permeable membrane, and nitrate and other ions are rejected. The driving force in reverse osmosis is pressure that exceeds the solution’s typical osmotic pressure.

iii) **Ion Exchange**

Ion Exchange introduces another substance, normally chloride, to substitute nitrate in the water. The ion exchange system is a tank filled with special resin beds that are charged with chloride. [10] As contaminated water flows through the tank, the nitrate exchanges with the chlorides. The resin can be recharged by backwashing with a sodium chloride solution. This method is very effective unless the water contains high amounts of sulfate, which may compete with nitrate in the exchange process. In addition, the contaminated water must go through a neutralizing process prior to treatment. Finally, the backwash brines, which are high in nitrate, must be disposed of properly. An ion exchange unit can cost 18000/- to over 154000/- for large models. Ion exchange is a process in which the target ion replaces another less well adsorbed ion on a resin. This process is a promising technology for nitrate removal because of its simplicity, effectiveness, and relatively low cost.

iv) **Electrodialysis**

It is a combination of reverse osmosis and ion exchange techniques. The driving force across the membrane is provided by electric current. The ions are removed from the water and pass through the membrane, attracted by the opposite electric charge on the other side of the membrane. [10] The advantages of the system are that the residence time controls the amount of dissolved solids removed, and that the system can be run continuously with no regeneration required. However, in the process, water must carry an electric current, and as the water is cleaner it offers more resistance to the current, that increases the cost of operation. The process may also be applied in the soil while the movement of water solution through profile before it reaches to ground water. It is found that the elecrot migration is an effective means for concentrating and retaining nitrates close to the anode in the saturated soil at low flow rates. For a given electrical input, the effect was reduced as the hydraulic flow rate increased.

v) **Blending**

High-nitrate groundwater is largely managed by blending the groundwater with surface water that has lower nitrate concentrations. This approach is common for drinking water treatment in larger municipalities and obviously requires a second, low-nitrate water source. [10] For example, in country like central Arizona, high-nitrate groundwater is pumped into canals conveying surface water to blend the nitrate level to well below the permissible level. But contaminated groundwater, agricultural drainage waters, and municipal wastewater, once considered unusable, are increasingly being seriously considered as sources of municipal water in order to meet current and future drinking-water demands. Thus removing nitrate and other contaminants will become more critical.

vi) **Chemical Denitrification**

Another technology for nitrate removal is chemical denitrification in which iron or aluminum is used to reduce nitrate to ammonia or nitrogen gas. Metals such as platinum, palladium, tin, and copper can chemically reduce nitrate to other forms, but they usually require a low pH, often need the addition of hydrogen gas or another strong reductant, and perform best with added heat. [10] As a result, full-scale treatment systems based on these catalysts are not yet used for drinking-water applications. Zero-valent iron (Fe0) has gained the most attention as a nitrate reductant system. Both in-situ groundwater and above-ground treatment systems have been demonstrated at several sites and commercial vendors have recently entered the marketplace. Oxidation of the
iron frees electrons, which are then available for nitrate reduction. The precise reactions for zero-valent iron and other chemical reduction processes are not well known for groundwater matrices, but in most cases nitrate reduction in groundwater does not proceed to innocuous gases. Instead, the majority of the nitrogen transforms into ammonia, which poses other water-quality challenges that may necessitate further treatment. Once researchers discover how to force the reaction to nitrogen gas, wide scale applications of these technologies may be developed for above-ground treatment systems.

vii) Biological Denitrification

Many bacteria belonging to different genera can grow anaerobically by reducing ionic nitrogenous oxides to gaseous products. Nitrates or nitrites served as the terminal electron acceptors instead of oxygen and resulted in generation of Nitrogen Gas. Such denitrification was dissimilatory nitrate reduction. When electrons are transferred from the donor to the acceptor, the organism gains energy, it is applied for the synthesis of a new cell mass and the maintenance of the existing cellmass. The enzymes associated with denitrification are synthesized under anaerobic or partially aerobic conditions. Nitrate reduction to nitrogen gas occurred as: \( \text{NO}_3^- \rightarrow \text{NO}_2^- \rightarrow \text{NO} \rightarrow \text{N}_2 \text{O} \rightarrow \text{N}_2 \)

Each step was catalyzed by an enzyme system. Dissimilatory reduction of nitrate to nitrite was important for most bacteria, since the process involved energy conservation by increased substrate level phosphorylation reaction. Since denitrification was a respiratory process, an oxidisable substrate was needed as an energy source. In biological denitrification there is possible bacterial contamination and presence of residual organics which can be overcome by disinfection treatment.

Nitrate contamination in groundwater is one of the emerging issues of drinking water quality in our country although it is established now that the low cost filters and best available technology can be effectively used for the treatment of nitrate contaminated waters. But the problem doesn’t end with mitigation as it is an end-of-pipe solution and the technology such as Reverse osmosis, Ion exchange, Electro dialysis create a stream of concentrated nitrates and have disposal problems. Most nitrate treatment systems are geared toward treating groundwater, and often are operated only periodically to meet site-specific, often seasonal water demands. Reverse osmosis, elect dialysis, and ion exchange require an input of electrical power or regeneration chemicals and also produce waste streams with high levels of nitrate and other salts.

I. Advances in Nitrate Removal

At high nitrate concentrations, water must be treated to meet regulated concentrations. But, it is almost impossible to remove nitrate by conventional drinking water treatment methods such as coagulation and filtration due to its high stability and solubility, as well as its low potential for co-precipitation or adsorption in water. Therefore, other technologies including such as ion exchange, reverse osmosis, electrodialysis, and chemical denitrification have been studied or applied to remove nitrate from drinking water. World Health Organization has suggested Ion Exchange as nitrate removal methods but biological denitrification can be future promising technology. Each of these technologies has its own strengths and drawbacks and their feasibility is weighted against factors such as cost, water quality improvement, residuals handling, and post-treatment requirements. While US EPA has suggested Ion Exchange, Reverse Osmosis requiring high TDS disposal and has a high operational cost and large volume of reject water, & Electrodialysis, as Best Available Technologies to treat nitrate contaminated water. Comparison of these in-home methods for removal of nitrate from drinking water is given in Table 1.

<table>
<thead>
<tr>
<th>Method</th>
<th>Initial Cost Rs.</th>
<th>Annual Running Cost</th>
<th>Maintenance</th>
<th>Speed</th>
<th>Remark</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Distillation</td>
<td>17500/- to 105000/-</td>
<td>28000/- to 35000/ + electricity</td>
<td>Periodic scale removal</td>
<td>15 to 38/Liter/day</td>
<td>Simpler, easier to maintain, Higher electricity costs</td>
</tr>
<tr>
<td>2 Reverse Osmosis</td>
<td>21000/- to 91000/-</td>
<td>7000/- to 21000/ + electricity</td>
<td>Filter replacement</td>
<td>7 to 38/Liter/day</td>
<td>Wastewater loss, High maintenance</td>
</tr>
<tr>
<td>3 Ion Exchange</td>
<td>18000/- to 154000/-</td>
<td>9000/- to 65000/ + electricity</td>
<td>Disposal of blackwash brines</td>
<td>Only limited to size of tank</td>
<td>Sodium Content in water may be health risk to humans and plants</td>
</tr>
</tbody>
</table>

There are many advances in nitrogen removal in last decade but most of the advances are for wastewater nitrogen removal. Increased need of recycling of wastewater for further reuse promoted large scale treatment of wastewater. As there are more contaminants in wastewater it requires primary, secondary and tertiary treatment for polishing of treated water. In most of the tertiary treatment various ways of biological nutrient removal techniques are used. Few of the recent developments in biological nutrient removal and advances in wastewater nitrogen removal by biological process are briefly described below.

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Activated Carbon is considered to be a universal adsorbent and it removes diverse types of aquatic pollutants. Powdered activated carbon and carbon nanotubes were also used for the removal of NO$_3^-$, where removal capacity of Carbon Nanotubes was found to be higher than powdered activated carbon. Granular activated carbon obtained from coconut shells have also been examined for NO$_3^-$ removal, using steam activation process. Clay being a natural scavenger can also be used for NO$_3^-$ removal. Calcium bentonite and Calcium Montmorillonite have been used and carbon nanotubes showed better removal capacity. Use of agricultural waste material is another option and various waste materials like wheat straw, raw wheat residue, and sugarcane bagasse and rice hull being used for effective NO$_3^-$ removal from water. In addition to the above there are other methods for removal of dissolved salts from the water such as, freeze desalination, liquid/liquid extraction, etc. The feasibility and economics in removing nitrates from the ground water should have been evaluated.

Biological nitrogen and phosphorous removal from municipal wastewater with the activated sludge system has been the preferred technology for the last 40 year. While several questions remains to be answered for more consistent reliable and stable performance for enhanced biological phosphorus removal, recent developments in this technology have focused on (i) increasing capacity and reducing the plant space and (ii) improving Nitrogen removal. To increase capacity and reduce Activated Sludge system space (a) integrated fixed film activated sludge, (b) external nitrification, (c) membrane, (d) aerobic granulation biological nitrate removal systems and (e) more efficient Nitrate removal bioprocesses have been developed.\(^8\)

With Integrated fixed film activated sludge system, fixed media are added to aerobic activated sludge reactor to make nitrification independent of the suspended activated sludge age. With External nitrification, nitrification is achieved in a side-stream fixed media reactor, which removes the size defining nitrification process from the suspended activated sludge reactor and halves its sludge age, improves sludge settleability and increase capacity. With membranes, secondary settling tanks are replaced with in-reactor membrane for solid – liquid separation. With aerobic granulation, the activated sludge process is controlled to form fast settling granules comprising heterotrophs, nitrifiers, denitrifiers and phosphorous accumulating organisms in a sequencing batch type reactor. The granules not only settle fast but the SBR type operation also removes the need for secondary settling tanks allowing higher reactor solids concentrations and hence smaller reactors.\(^7\) To achieve Nitrate removal more efficiently, methods are being developed to (i) short circuit nitrification denitrification by preventing nitrate formation and enforcing Nitrification – denitrification over nitrite. This requires less oxygen and organics than Nitrification – denitrification over nitrate allowing lower Nitrate concentration to be achieved for the same influent organic concentration and oxygen supply and (ii) encouraging the growth of annammox bacteria in the activated sludge which removes Nitrate autotrophically by combining ammonia and nitrite to form nitrogen gas. This halves oxygen demand for nitrification and requires no organics. But all these recent development in Biological nitrogen removal technology are not applied in treatment of drinking water.

Apart from common technology discussed above i.e. distillation, blending, reverse osmosis, ion exchange, electrodialysis and chemical denitrification for nitrate removal from water, there are few advance technologies used by some researchers for nitrate removal. These advanced technologies are Catalytic denitrification, Electro catalytic reduction for denitrification, Denitrification using a membrane bioreactor, combined ion exchange & membrane bioreactor for denitrification, Denitrification using nanofiltration and biological denitrification. Brief of these technologies is given below.

i. Catalytic Denitrification

A catalytic process was developed for the removal of nitrite and nitrate from water. Palladium - alumina catalysts were effective in reducing nitrite to nitrogen (98%) and ammonia in the presence of hydrogen. The lead (5%), copper (1.25%), Al$_2$O$_3$, catalyst were found to completely remove nitrate from water having an initial nitrate concentration of 100 mg/L. The reaction was completed in 50 min. The process operated effectively at a temperature of 10°C and pH 6-8.\(^6\)

ii. Electro catalytic Reduction for Denitrification

An electrocatalytic reduction process was used to remove NO$_3^-$ from groundwater. A commercially available carbon cloth with a 30% surface coated Rh (rhodium) (1 μg/cm) was tested at an applied potential of −1.5 V versus standard calomel electrode (SCE) with a Pt auxiliary electrode. The results suggested that electrocatalysis reduced NO$_3^-$ concentrations in groundwater from 73 to 39 mg/L on a timescale range of 40-60 min.\(^6\)

iii. Denitrification using a Membrane Bioreactor

Immersed heterotrophic membrane bioreactor (MBR) produced high quality product water34 when NO$_3^-$ contaminated water was made to flow through the lumen of tubular microporous membranes. NO$_3^-$ diffused through the membrane pores. Denitrification took place on the shell side of the membranes35. The MBR achieved over 99% NO$_3^-$ removal at an influent concentration of 200 mg NO$_3^-$ per liter.\(^6\)

iv. Combined Ion Exchange & Membrane Bioreactor for Denitrification

The IEMB concept combines dialysis and simultaneous biological degradation of nitrate in small concentrations. The IEMB process operated with hydraulic retention times ranging from 1.4 to 8.3 h in the water
compartment, proved to remove nitrates effectively, while preserving the water composition with respect to other ions, thus avoiding secondary contamination of the treated water.\(^6\)

v. Denitrification using Nanofiltration

During the last decade, nano filtration made a breakthrough in drinking water production for the removal of nitrate. For the removal of nitrates the membranes NF70, NF45, UTC-20, and UTC-60 have been experimentally studied. The results showed that only a small fraction of nitrate was removed for most membranes, except for NF70 where a 76% nitrate removal was obtained.\(^6\)

There are many emerging technologies where in Denitrification process is used for nitrate removal from wastewater in many researched by some of agencies and patented their process. ANAMMOX granulated sludge blanket reactor developed by Paques Company, China, DEMON Sequencing batch reactor developed in USA, DeAmmon moving bed biofilm reactor developed by Lackerby in Sweden, ANITA Mox mbbr system developed by Anox Kaldnes, Terra-N Process developed by Claritant in Munich Germany, are emerging technologies based on Denitrification.\(^6\) But all these processes are mostly designed for wastewater. Based on above technologies Biological Denitrification for drinking water nitrate removal can be designed.

Although Biological Denitrification process is commonly applied in wastewater treatment, its application for drinking and groundwater treatment has not investigated in lab studies and only rare developed in full-scale plants. However, potential contamination of the treated water with these microorganisms and their metabolic byproducts are the drawbacks of this technology. These problems result in increased disinfectant demand or the need of post-treatment of the product water by filtration. In addition, low production rates and cold temperature restrictions can also be considered as a disadvantage of biological denitrification. Biological denitrification is widely used for the treatment of municipal and industrial wastewater by degradation of microorganisms, but is less commonly used in drinking water applications.

Conclusions

Removal of nitrates from drinking water is an important and developing area of research. Technology development has occurred in this area, but further optimization of current technologies is required. The overall goal of this paper was to explore various practical and economical way to reduce nitrate concentrations in groundwater. This can be achieved by conducting an experimental trial by developing a laboratory procedure to test the effects of difference conditions on denitrification. From the above short review, it is clear that biological removal of nitrate from groundwater is a good solution in many aspects. To utilize the opportunity, research work can be carried out to design and optimize the small scale mixed bed bio reactor technology for biological denitrification of ground water to be supplied for village.

RO and ED performance data for nitrate removal is limited and the impact of rapid advances in these technologies should be examined. Biological denitrification is one of the most effective technologies for nitrate removal since it only removes nitrate and doesn’t change concentrations of other background ions and nitrate is microbially reduced to nitrogen gas. Biological denitrification reactor operation in regard to microbiological characteristics of biologically denitrified water demands attention.

Further research can be devoted for the contribution in the improvement of water quality in wells using mixed bed reactors/ filters to remove nitrate from drinking water. Biological removal of nitrate from drinking water can be studied in a mixed bed reactor like slow sand filter with using different medium had different particle size. Sand, gravel pack, and granite gravel can be used with adding ethanol or other ecofriendly material as source of carbon to enhancing the potential of denitrification. The flow rate, nitrate concentration and pH needed to be studied for their impact on nitrate removal through the sand filter. Such biological denitrification treatment systems need the addition of an electron-donor source, and the effluent from these systems contains biomass and potentially residual organics. Furthermore, most biological systems require time for biological acclimation and so cannot be turned on or off as needed. Despite the development of these technologies, a need remains for a low-cost, low-maintenance, efficient method to treat nitrate-contaminated groundwater.


