Some of the Innovative Uses of Waste Materials in Groundwater Treatment: A Mini-Review of Nitrate Removal with Recycled Rubber

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Some of the Innovative Uses of Waste Materials in Groundwater Treatment: A Mini-Review of Nitrate Removal with Recycled Rubber

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ABSTARCT

One of the most urgent environmental issues is groundwater pollution, especially in relation to nitrate removal, which presents significant dangers to human health and the environment. In order to address nitrate contamination in groundwater, this review examines creative and sustainable methods that make use of waste materials, specifically recycled rubber from discarded tires. Based on experimental and field performance, the article covers many approaches that have been developed employing recycled rubber as an adsorbent for nitrate and evaluates their efficacy. It also emphasizes the chemical and physical characteristics of recycled rubber, such as particle size, surface changes, and interaction parameters, that improve its effectiveness in removing nitrate. With the help of this thorough analysis, the study offers recommendations for improving recycled rubber's performance as a long-term and practical remedy for nitrate contamination. It also calls for more investigation and development to increase the system's effectiveness in a range of groundwater conditions.

1. Introduction

Groundwater nitrate contamination has been one of the unsolved issues with environmental policy for the past 30 years. Many drinking water production sites were threatened at once when the nitrate standard in the drinking water supply was reduced in 19861 from 90 mg/l to 50 mg/l. This was because the standard had either been reached, exceeded, or would be exceeded in the near future due to rising concentrations (Haakh, 2017). A valuable natural resource with significant social and economic implications is groundwater. Nearly half of the world's drinking water comes from groundwater, which is also essential to food production since it provides more than 40% of the world's consumptive usage of irrigation for agriculture. Additionally, from an environmental perspective, wetlands, lakes, and rivers are directly supported by groundwater supplies. Groundwater resources have been under increasing strain worldwide over the past few decades, which has frequently resulted in abstraction above sustainable levels and higher contamination levels. Global groundwater

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resources are facing many risks due to climate change, land use, and population increase. These factors are affecting the amount and quality of groundwater resources (Zhou et al., 2015). Groundwater is the only source of potable water for both urban and rural regions in many parts of the world. However, growing industrial and agricultural activity in recent years has led to the creation of harmful contaminants such metal ions, synthetic organic compounds, and inorganic anions, which has raised public concerns about the quality of groundwater. Since inorganic anions are poisonous and detrimental to both people and animals at very low quantities (ppb), they are extremely important. Since the existence of tiny amounts of hazardous inorganic anions typically does not cause organoleptic changes in drinking water, some of them may go undiscovered, raising the potential hazards to human health (Bhatnagar and Sillanpää, 2011). A hazard to aquatic ecosystems and humanity is the contamination of surface and groundwater. Providing clean water for human use, food production, and recreational purposes is unquestionably one of the most urgent challenges of the twenty-first century. Freshwater availability has significantly decreased due to global warming, desertification, and contamination of surface and groundwater. This is a major problem for achieving the Sustainable Development Goals (SDG) in the 2030 Agenda for Sustainable Development, which was endorsed by all UN Member States (Fernández-López et al., 2023). The most important anthropogenic sources of groundwater nitrate pollution are most likely agricultural practices. Adults who use drinking water with high nitrate contents may develop stomach cancer or methemoglobinemia in new-borns. An essential ingredient for promoting plant development is nitrogen. This fact has led to the widespread application of nitrogen-based fertilizers to increase agricultural output throughout a large portion of the globe (Almasri and Kaluarachchi, 2007). Concern over nitrate (NO_3^-) pollution of surface and groundwater bodies is growing worldwide, and this has sparked a great deal of scientific interest. Since nitrogen is a macronutrient that is essential to all life on Earth, increased human activity has significantly changed the global nitrogen cycle, causing eutrophication and hypoxic conditions in aquatic environments. Intense agricultural methods produce massive nitrate fluxes that unintentionally worsen water quality because of nitrogen over fertilization. Additional industrial activities add to the environmental contamination caused by NO_3^-. Various methods are available to effectively achieve denitrification of waterbodies, guaranteeing acceptable amounts of NO_3^- (Almasri and Kaluarachchi, 2007). The use of nitrogenous fertilizers has expanded, land use patterns have changed, and residential wastewater recycling has increased, all of which have contributed to an increase in nitrate levels. Even while nitrate is thought to be reasonably safe for adults, in new-borns, it is reduced from NO_3^- to NO_2^-, which then binds with haemoglobin in the blood to produce methamoglobin, causing the disease that is frequently referred to as "blue baby syndrome." A 45 mgNO_3^-/ L limit was set by the Health and Welfare World Guidelines (Archna et al., 2012). Methaemoglobinemia in infants and, at higher levels, cattle poisoning is caused by excessive nitrate levels in drinking water. Both have the potential to be lethal (Tredoux and Talma, 2006). Since nitrate is so common and so hazardous, it is considered a priority contaminant of groundwater in many nations. The usage of nitrogen fertilizers and irrigation with home wastewater are the primary causes of nitrate contamination. As a result, nitrate pollution is present in both industrialized and developing nations. Twenty percent and forty percent, respectively, of the EU's groundwater quality monitoring stations had concentrations of more than 50 mg and 25 mg of NO3/l between 1996 and 1998; in contrast, several poor nations had high concentrations.

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Additionally, nitrate concentrations were noted (Della Rocca et al., 2007). A multitude of intricate processes, including soil nitrogen dynamics, on-ground nitrogen loading, groundwater recharge, soil properties, and the depth to the water table, combine to cause nitrate leaching from the unsaturated zone (Almasri, 2007). Agricultural farming encompasses a wide range of operations, from large-scale irrigation projects to intense animal feeding operations and substantial dry land grazing. There are big differences in these activities' potential for nitrate contamination (Tredoux and Talma, 2006).

Diffuse pollution is the problem with nitrate pollution because it is created by the use of manure, excessive use of nitrogen fertilizers, and crop irrigation using household wastewater. Applying the ex-situ method to groundwater clean-up is extremely challenging due to non-point sources of nitrate pollution. However, due to their technological complexity, the best existing methods for treating nitrate-contaminated water, such as reverse osmosis, ion exchange, and electro dialysis, cannot be employed in-situ for the generation of drinking water (Della Rocca et al., 2007).

These days, garbage need to be viewed as a resource and sustainability is a major concern. Because of this, a substantial body of research is being done on the topic of using sustainable methods to handle the massive amount of post-consumer tires. Tire research is still going strong, with the goal of developing environmentally friendly solutions that would both minimize waste and maximize the use of existing technology and processes to produce "green" tires that can be widely recycled (Battista et al., 2021). For drinking water, the World Health Organization has set a maximum of 10 mg $NO3^-$ N/L. A thorough framework for preventing NO3 contamination of waterways was built in Europe with the introduction of the Nitrate Directive. However, a deeper understanding of NO3 source inputs is necessary to regulate NO3 pollution in water efficiently. Afterwards, specific actions might be taken to reduce or stop contamination (Xue et al., 2009).

This review article's primary goal is to offer a thorough assessment of the creative application of recycled rubber from used tires as an adsorbent for the removal of nitrate in groundwater treatment. The article aims to evaluate recycled rubber's efficacy, mechanisms, and potential as a long-term remedy for nitrate contamination by methodically examining the literature. It also highlights the advantages and disadvantages of this approach from an environmental, financial, and technical standpoint. The assessment also attempts to pinpoint areas of unmet research need and potential future directions for enhancing the functionality and scalability of recycled rubber in groundwater treatment systems.

2. Groundwater Contamination by Nitrates

Groundwater is an essential component of the region's overall hydrologic cycle. One of the main channels via which groundwater pollutants interact with people and the larger terrestrial environment is through interactions between groundwater and surface water bodies (recharge and discharge zones). Due to the polluted groundwater being diluted, these interactions may be advantageous. This may play a significant role in lessening the effects of contaminated groundwater. Alternatively, pollution may be absorbed and stored in plants and animals, or it may get concentrated in bottom sediments due to sorption and precipitation processes (Hashim et al., 2011).

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Because nitrate is very soluble in water and does not attach itself to soil easily, it is quite prone to leaching. Animal wastes, septic tanks, municipal wastewater treatment facilities, and decomposing plant detritus are a few possible sources of nitrate. Nonetheless, it is thought that the primary cause of nitrate contamination in the environment is agricultural fertilizers that are enhanced with nitrogen. The US EPA and WHO have set a nitrate limit for drinking water of 50 mg/L nitrate-NO3 or 10 mg/L Nitrate-N in order to lessen the health risk. Nonetheless, reports have indicated that in many regions of the world, the quantity of nitrate in drinking water has beyond the maximum allowable level. Controlling the nitrate level in drinkable water below the recommended threshold has therefore grown to be of great significance. For example, it was projected that by 2010, the UK government was receiving payments of around £ G58 million year to maintain the quantity of nitrate in drinking water below the permitted limit (Mohsenipour et al., 2014). There are several methods and causes of nitrate pollution in groundwater systems. Addressing issues with groundwater quality requires an understanding of system dynamics and the identification of the several sources of nitrate pollution. The two primary categories of nitrate pollution sources are generally nonpoint (diffuse) and point-source pollution. The main nonpoint source of contamination impacting groundwater quality is the use of fertilizers in agriculture. Unlike point sources, which are distinct, isolated sources of pollution that primarily affect limited areas, this type of pollution is widespread. Long-term, widespread abuse of chemical or manure fertilizers (on lawns, golf courses, and crops) as well as persistent sewage line leaks are examples of diffuse sources of nitrate (Zhou et al., 2015).

3. Recycled Rubber as an Adsorbent

According to WHO and EPA criteria, adsorption technology has advanced over the past ten years to become an effective and ubiquitous technique of treating water. Its affordability and environmental friendliness account for its broader use and increased momentum. Because this method uses efficient adsorbents made from solid industrial, agricultural, and municipal wastes, it is very cost-effective. Its need has been further increased by the conversion of such low-value solid waste into value-added goods with applications in the environmental field. The creation of more effective adsorbents from solid waste products is still being researched (Gupta et al., 2012).

Over 10 billion tires are thought to be thrown annually worldwide; in the US, over 259 million tires were produced in 2005. About 80% of worn tires are used in the scrap tire market, with the other 20% being stacked up or dumped in landfills. Tire crumb rubber (TCR) is made up of a complex blend of elastomers, such as styrene-butadiene, polybutadiene, and polyisoprene. Additional significant ingredients in tires are stearic acid (1.2%), zinc oxide (1.9%), extender oil (1.9%), and carbon black (31.0%). Rubber is strengthened, its resistance to abrasion is increased, and UV ray deterioration is decreased with the application of carbon black (CB). This nanoscale component ought to have adsorption qualities akin to activated charcoal, a well-known removal agent for organic compounds (Alamo-Nole et al., 2011). Conversely, a buildup of trash tires presents major health and safety risks. Entire tires function as havens for rats and insects that spread illness. Tire piles that are out of control can burn for months, releasing dangerous oil residue and bitter black smoke. Tire piles

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are also fire dangers. The same issues that arise with hoarding also arise with widespread unlawful dumping. In addition to being a major environmental issue, the vast quantity of discarded tires may be used to create inexpensive adsorbent materials that might be used to remove heavy metals from solutions (Calisir et al., 2009).

There are several previous studies and research that have utilized recycled tire rubber as an adsorbent material for various pollutants. Some of these studies include: In a study by (Calisir et al., 2009), crumb rubber was investigated as an adsorbent for the removal of Cu(II) ions from aqueous solutions. The study examined the effects of various factors, including pH (ranging from 1.5 to 7.0), contact time (6 to 96 hours), and initial copper concentrations (1 to 50 mg/L). The findings indicated that Cu(II) adsorption is highly pH-dependent, with optimal removal occurring at pH 6.0. The copper uptake was also associated with the displacement of zinc, suggesting an ion exchange mechanism. The Langmuir and Freundlich adsorption models were applied to describe the adsorption isotherms, with equilibrium data aligning well with the Langmuir model. Overall, the study demonstrated that crumb rubber is an effective adsorbent for Cu(II) ion removal from aqueous solutions.

A novel carbon (RTAC), developed from waste tire rubber via physical activation, was evaluated as an adsorbent for the removal of lead (Pb^{2+}) and nickel (Ni^{2+}) ions from aqueous solutions in a study achieved by (Gupta et al., 2012). The mesoporous structure of RTAC enhanced its adsorption capacity compared to a commercial microporous carbon (CAC), showing a higher affinity for Pb^{2+} than Ni^{2+} due to differences in electronegativity and ionic radii. Various operating parameters were examined, and equilibrium data fit the Langmuir model, while kinetic data followed the pseudo-second-order model. The adsorption process involved initial surface adsorption and intraparticle diffusion, with thermodynamic studies confirming the system's feasibility and endothermic nature. Column experiments demonstrated effective lead (96%) and nickel (87%) removal from simulated electroplating wastewater, and the system proved reusable with minimal loss of efficiency. These findings highlight the potential of RTAC as an economically viable, eco-friendly adsorbent for wastewater treatment, offering technical feasibility and easy synthesis.

The adsorption of phenol from aqueous solution using waste tire rubber granules (WTRG) was studied in a batch system by (Aisien et al., 2013). Various factors affecting adsorption capacity were examined, including contact time, phenol concentration, adsorbent amount, particle size, pH, and temperature. Equilibrium data were analyzed using Langmuir and Freundlich isotherm models, while the kinetics were studied with Lagergren pseudo-first-order, pseudo-second-order, and intra-particle diffusion models. The equilibrium time for adsorption was 60 minutes, indicating rapid adsorption. Adsorption capacity increased with smaller particle size and lower solution temperature, with maximum adsorption at pH 8.5. The Langmuir isotherm best fit the equilibrium data (R^2 = 0.993), and the Lagergren pseudo-first-order model provided the best kinetic fit.

The study done by (Aliyu et al., 2022) achieved 90% tetracycline (TC) removal under optimal conditions of 30 mg of modified rubber waste (MRW), 30 mg/L TC concentration, pH 3, and 25°C after 5 hours. Adsorption data followed the Langmuir isotherm, indicating monolayer adsorption with a maximum capacity of 76.33 mg/g, and aligned with the pseudo-second-order kinetic model. Thermodynamic analysis showed the process was exothermic and spontaneous. After ten adsorption-desorption cycles, MRW maintained 90% efficiency. MRW also exhibited strong adsorption capacity

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for removing TC from municipal wastewater and tap water, suggesting its potential as a low-cost adsorbent for pharmaceutical removal from water and wastewater.

4. Mechanisms of Nitrate Adsorption

Nitrate ions (NO_3^-) are exchanged with other anions that are present on the rubber's surface as part of the ion exchange process. The negatively charged nitrate ions in polluted water can displace other anions (such sulfate or chloride) that may be weakly bonded to the rubber surface when recycled rubber comes into contact with it. When the rubber contains functional groups that may interact with the nitrate ions, this exchange mechanism is frequently preferred. As a result, the amount of nitrate ions in the rubber matrix increases, while the amount of nitrate in the water decreases accordingly (Zedler et al., 2022).

Weak van der Waals forces are the mechanism underlying the physical adsorption (physisorption) process, which draws nitrate ions to the rubber's surface. When nitrate ions are drawn to the rubber's surface by intermolecular interactions, physisorption takes place. This reaction is usually reversible and doesn't require any strong chemical bonds. The rubber's porosity and surface area determine how strong the adsorption is. Increased surface area and more porosity can improve the adsorption capability (Yu et al., 2022).

While stronger chemical bonds are formed between the adsorbate (nitrate ions) and the adsorbent (recycled rubber) in chemisorption, as opposed to physisorption, in chemical adsorption. This may happen if the rubber has functional groups (such as hydroxyl groups, carboxylic acids, or amines) that can react chemically with the nitrate ions to produce ionic or covalent connections. This procedure, which is often irreversible, helps to remove nitrate ions from water in general (Wang et al., 2021).

The creation of surface complexes between nitrate ions and the rubber's surface functional groups is a component of the surface complexation process. Surface complexes can occur as a result of interactions between nitrate ions and the functional groups on the rubber surface. By altering the electrical environments of the rubber and the nitrate ions, this interaction can improve adsorption. The availability of functional groups on the rubber is affected by this process, which is determined by the pH and ionic strength of the solution (Adusei-Gyamfi et al., 2019). Some of the mentioned technologies are illustrated in Fig.1.

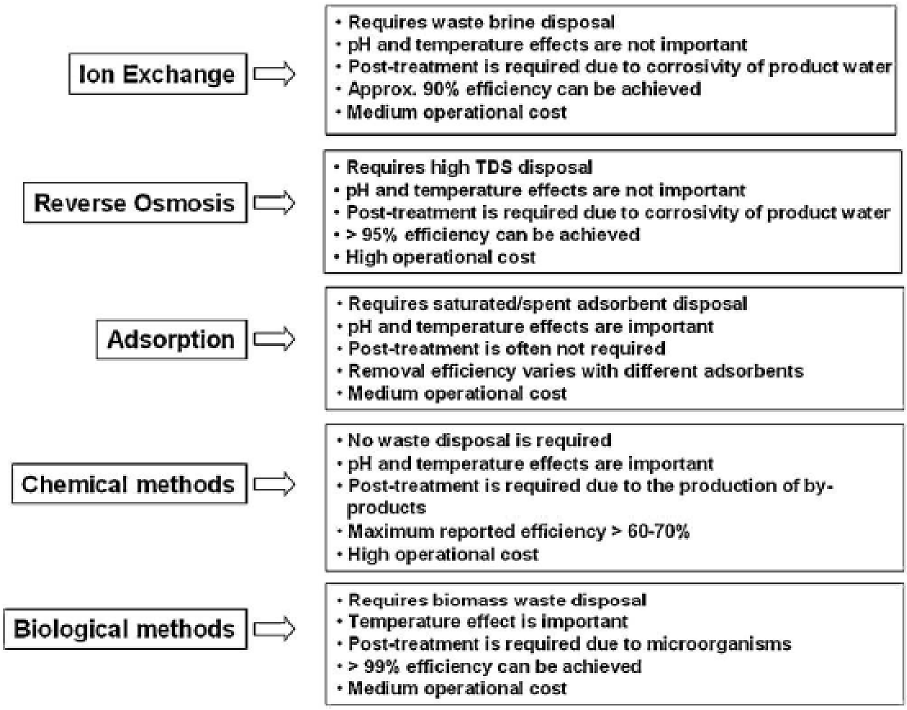

Fig. 1: Comparison of some nitrate removal technologies (Bhatnagar and Sillanpää, 2011)

5. Performance of Recycled Rubber in Nitrate Removal of Nitrate

Numerous earlier research has shown that recycled rubber, especially pulverized tire rubber, is a highly effective adsorbent for removing nitrate from aqueous solutions. Ground tire rubber that has been changed by heating it in a microwave using Wijs' reagent and polyamines has a far higher capacity to adsorb nitrate than untreated rubber. For example, PHA-300 W, a rubber modified with polyamines, had the maximum adsorption capacity of 3.61 mg/g; DTA-450 W and EDA-300 W had capacities of 3.08 mg/g and 2.16 mg/g, respectively (Rungrodnimitchai and Kotatha, 2019). A monolayer adsorption on a surface with a finite number of identical sites is suggested by isotherm studies, which showed that the adsorption behavior fits well with the Langmuir isotherm model (Chyan et al., 2013). The effective integration of polyamines into the rubber structure, which improved its adsorptive qualities, was further validated by elemental analysis and Fourier-transform infrared spectroscopy (FT-IR). Images obtained using scanning electron microscopy (SEM) showed that the modified rubber's particle sizes had shrunk, indicating structural alterations brought about by the aminolysis process. These modifications may have improved the material's adsorption capability by increasing its surface area and porosity. Apart from the results obtained in lab settings, one novel way to improve nitrate removal in artificial wetland systems is to employ waste rubber tire chips (WRTCs). Achieving an average removal rate of 62.2% as opposed to 13.1% in conventional gravel beds, hybrid systems that combine subsurface flow (SSF) built wetlands with WRTCs as the medium have shown impressive nitrate removal efficiency. Additionally, having WRTCs increased overall wastewater treatment (Saleh et al., 2013). Additionally, nitrate removal efficiencies have been shown

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to be significantly improved by using waste rubber tire chips (WRTCs) in engineered wetlands. In hybrid systems, removal rates have been shown to reach as high as 62.2%, whereas typical gravel beds only obtain 13.1%. This research implies that WRTCs improve wastewater treatment systems' overall efficiency in addition to acting as efficient adsorbents (Yeh and Wu, 2009).

6. Challenges and Opportunities in the Application of Recycled Rubber

Since the Industrial Revolution, rubber—also known as vulcanized rubber or cured rubber compound—has been utilized in many different industrial applications. Specifically, the advancement of the vulcanization process made it possible to produce huge quantities of high-quality rubber at a reasonable cost. Currently, 26.7 million tons of rubber are produced worldwide, of which 12.31 million are natural and 14.46 million are synthetic. These rubbers are used to make tires and other consumer and industrial goods. An estimated 1.5 billion tires are produced worldwide each year, and roughly the same amount of tires reach the end of their useful lives each year (Medina et al., 2018) as seen in Fig.2.

Used tire recycling now starts with the mechanical breakdown of waste tires by appropriate grinding, pulverization, and shredding. The beginning material for subsequent applications, which rely heavily on the dimensions, surface characteristics, and purity of the obtained ground tire rubber (GTR), is obtained GTR. The average and distribution sizes of the particles have a significant impact on the ultimate cost of rubber granules. This cost is directly connected to the energy used in the grinding, pulverization, and shredding of discarded tires (Formela, 2022).

The challenge of minimizing effects during the course of the product's life cycle must be addressed in the quest for new environmentally friendly materials and technologies. At this stage, polymeric materials have a number of benefits over other material classes, including low density, low processing temperatures, high specific resistance, and affordability. However, polymers are now frequently referred to as environmental villains, mostly because to the rapid growth in manufacturing and improper disposal of these materials in recent years.
Rubber waste recycling has some characteristics when it comes to polymeric material classes that aren't present in thermoplastics (de Sousa et al., 2019).

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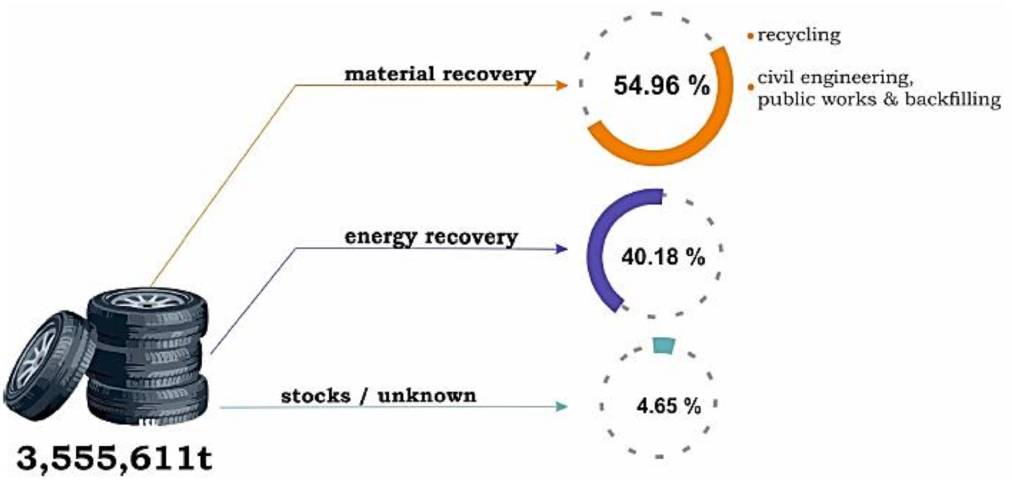

Fig. 2: Used tires management in Europe (Czarna-Juszkiewicz et al., 2023)

7. Conclusions

Contamination of groundwater with nitrates poses a serious threat to aquatic ecosystems and human health. The main causes of this problem are the widespread use of nitrogen-based fertilizers and agricultural methods that have raised drinking water nitrate levels over the permissible limits set by health authorities. Since groundwater provides about half of the world's drinking water, nitrate contamination has serious consequences that call for immediate action to protect this vital resource.

The novel potential of using recycled rubber from old tires as a powerful adsorbent for nitrate removal in groundwater treatment is highlighted in this paper. Studies show that recycled tire rubber exhibits a significant ability to adsorb nitrate via a number of different processes, such as physisorption, chemisorption, and ion exchange. Rubber may be modified by adding functional groups, which improves its adsorptive qualities and raises the efficiency of removing nitrates from the material.

Additionally, recycling used tires offers a sustainable solution to groundwater nitrate contamination while also addressing the environmental issues related to tire disposal. Even with encouraging outcomes, further research is required to maximize recycled rubber's performance, look into its long-term efficacy, and assess its scalability for wider groundwater remediation applications.

To sum up, using recycled rubber to minimize nitrate pollution in groundwater is a practical, economical, and ecologically beneficial solution. The advancement of sustainable water treatment technologies and the accomplishment of the clean water and sanitation-related Sustainable Development Goals depend heavily on ongoing research and development in this area.

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