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Research progress in tracing the source of groundwater pollution and remediation technologies

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Abstract: The management of groundwater pollution is a significant issue for safeguarding people's right to healthy water usage and achieving sustainable social development. Identifying pollution sources is a fundamental measure to cut off pollution sources and achieve radical pollution control. Pollution remediation is crucial for reducing pollution hazards, mitigating pollution risks, and building a healthy water resource system. Currently, common methods for tracing groundwater pollution sources include Water Quality Analysis, Isotope Tracing, Fluorescence Spectroscopy, and others. The common groundwater remediation techniques are broadly categorized into Ex–situ and In–situ Remediation Technologies, encompassing various methods and techniques. Based on a review of the above important content, this paper proposes that the research on groundwater pollution analysis and treatment currently exhibits a trend of coupling multiple tools and methods, and big data technology will play a more significant role in groundwater pollution treatment. The treatment of groundwater pollution not only depends on the innovation of technical methods, but also relies on the promotion of water resource protection concepts and institutional safeguards. It is a major project related to national economy and people's livelihood.

Keywords: polluted groundwater remediation; pollution source tracing; sustainable development

1. Introduction

Groundwater is a valuable freshwater resource for humans. About 97% of the Earth's potentially available freshwater are stored as groundwater. For a long time, with the discharge of domestic sewage and industrial wastewater, as well as the disorderly use of agricultural irrigation, fertilizers, and pesticides, scarce groundwater resources have suffered severe pollution. According to the monitoring results of 10,242 shallow groundwater quality monitoring points published in the "*China's Ecological Environment Status Bulletin 2020*", only 22.7% of the monitoring points were rated as Class I–III water quality, with 33.7% as Class IV and 43.6% as Class V. Numerous studies and statistical data indicate that the groundwater pollution in China exhibits a worsening trend characterized by increasing severity, continuous expansion of contaminated areas, and growing diversity of pollution sources. Groundwater pollution poses a serious threat to the safety of drinking water, thereby affecting human health. In 2021, the State Council issued the "Regulations on the Management of Groundwater", emphasizing that pollution prevention and control are the top priorities in groundwater management. However, groundwater pollution is latent. Its management is a complex and systematic engineering that includes pollution source tracing and pollution remediation.

Scholars from both domestic and international communities have carried out fruitful explorations into the sources, properties, distributions and targeted remediation of groundwater pollutants. Groundwater pollution treatment should base on accurate identification of pollution sources, and consider the morphological distribution of pollutants and pollution diffusion processes. Therefore, this study systematically analyzes research progress in groundwater pollution treatment from the perspectives of groundwater pollution source tracing and groundwater pollution remediation, laying a theoretical foundation for further research on groundwater pollution control.

2. Source tracing of groundwater pollution

Source tracing of groundwater pollution is a prerequisite for groundwater pollution treatment. Currently widely–used methods for tracing pollution sources primarily include Water Quality Analysis, Isotope Tracing, Fluorescence Spectroscopy, *etc.*

2.1. Water quality analysis

Water quality analysis is a method for identifying pollution sources by detecting the physical and chemical indicators of contaminants in groundwater and comparing them with potential surrounding pollution sources. It includes two methods: Graphical water quality analysis and Quantitative water quality analysis. Graphical water quality analysis is an approach that qualitatively identifies groundwater pollution sources by transforming monitoring data into visual graphics through visualization techniques, while incorporating environmental background information and wastewater discharge data of the study area. Quantitative water quality analysis is a systematic analytical framework that employs quantitative tools including the Chemical Mass Balance (CMB) model, Positive Matrix Factorization (PMF) model, and multivariate statistical models to precisely quantify pollutant species, concentrations, sources, and migration–transformation processes in water bodies. Its core objectives are to achieve accurate pollution source tracing, contribution rate calculation, and quantitative risk assessment.While water quality analysis methods can effectively characterize pollutant sources and their migration–transformation processes with relatively minimal prior knowledge of pollutant source fingerprints, they are susceptible to subjective influences that may introduce significant errors in the analytical results (Yu Lu, Zheng Tianyuan, *et al.*, 2022).

2.2. Isotope tracing method

The isotopic composition of water bodies varies characteristically depending on their distinct origins. This method analyzes the isotopic signatures in water to trace pollutant sources and migration pathways, thereby enabling identification of contamination sources and assessment of pollution levels in groundwater systems. In studies on source tracing of groundwater pollution, isotope methods for nitrate, sulfate, metals, *etc.*, have been frequently used by scholars domestically and internationally. It is mainly applied to the source tracing of nitrogen and phosphorus pollution, metal pollution, and so on (He

Yunyong, *et al.*,2022). With the continuous advancement of isotope technology, isotope tracer techniques have been widely applied in the field of ecological and environmental monitoring. However, this method demands high accuracy in sample analysis and requires highly precise analytical instruments, which entail significant costs for instrument acquisition, personnel training, and facilities. These are the main challenges to the promotion of this technique.

2.3. Fluorescence spectroscopy method

This method enables qualitative identification and quantitative analysis of water pollutants by detecting characteristic differences in their fluorescence spectra, thereby facilitating pollution source identification and contribution rate calculation. Methods utilizing spectroscopic principles to analyze material structural characteristics and chemical compositions mainly include Three–dimensional fluorescence spectroscopy, Synchronous fluorescence spectroscopy, Time–resolved fluorescence spectroscopy, and so on (Chen Yunan, 2017).

Source tracing is the fundamental prerequisite and critical link in the prevention and control of groundwater pollution. Currently, the mainstream methods such as Water quality analysis, Isotope tracing, and Fluorescence spectroscopy all trace pollution sources by identifying similarities and correlations between the physical structures and chemical compositions of contaminants in polluted water and potential pollution sources in the surrounding environment. Corresponding source tracing technologies can be continuously updated based on the evolution of physical and chemical analysis methods.

3. Groundwater pollution remediation

Groundwater pollution remediation refers to the process of treating contaminated groundwater through physical, chemical, biological, or combined technological approaches to reduce or eliminate pollutant concentrations or transform them to acceptable levels. Its core objectives are to halt pollutant migration and restore the usability of groundwater resources to the greatest extent possible. Depending on whether the contaminated water source needs to be treated at a different location, groundwater pollution remediation can be divided into Ex–situ remediation technology and In–situ remediation technology.

3.1. Ex–situ remediation technology

Ex–situ remediation technologies involve extracting contaminated groundwater, removing contaminants through physical, chemical, or biological methods, and then recharge the treated water. This can reduce the risks such as land subsidence caused by the extensive extraction of groundwater. The main methods of this technology include Pump–and–treat technology and Multi–phase extraction technology. Despite the high cost, ex–situ remediation technology generally requires less time to achieve efficient contaminant cleanup, is easily monitored and achieves more uniformity, and after ex–situ treatment, decontaminated soils may be used for landscape purposes (Kuppusamy S, Palanisami T, Megharaj M, et al., 2016) .

3.1.1. Pump–and–treat technique

This method involves pumping out the contaminated groundwater, treating it to meet standards, and then recharging. It is an early and widely used groundwater pollution remediation technology and a representative of Ex–situ remediation technologies. This technique is suitable for sites with high concentrations of contaminants, large contamination areas, and deeply buried sites. It has advantages of high initial remediation efficiency, large treatment capacity, and simple operation. But it may experience a tailing effect where the conversion rate of contaminants into water decreases and a rebound effect where pollutant levels gradually rise after pumping stops in the later stage (Zhao Yongsheng, 2012). Additionally, Pump–and–treat technique has minimal effect on non–aqueous phase liquids and significantly disturbs the groundwater flow field and aquatic environment (Pu Min, 2017). According to worldwide statistics, between 2010 and 2017, the use of this technology in groundwater remediation sites declined from a peak of 55%–100% to 17%–26% (Zhang Li, *et al.*, 2023). In recent years, scholars have built mathematical models of contaminated remediation sites, used numerical simulation software to simulate contaminant plume distributions under different pumping schemes, and selected optimization algorithms to adjust well placement and pumping modes to find the optimal pumping plan (Zeng Xiankui, *et al.*, 2012).

3.1.2. Multi–phase extraction technology (MPE)

MPE is a remediation method that employs vacuum extraction to simultaneously remove and treat contaminated soil, polluted groundwater, non–aqueous phase liquids (NAPLs), and vapors from subsurface zones by extracting them to the surface for separation and treatment. It is specifically targeted at volatile and mobile non–aqueous phase liquids. This method can simultaneously remediate contaminants in groundwater, the vadose zone, and aquifer soils. It typically consists of a multi–phase extraction system, a multi–phase separation system, and a contaminant treatment system. MPE has a large remediation scope and can treat various types of water contaminants, including gaseous, liquid, and solid phases. It can also reduce the risk of groundwater re–contaminating the aquifer soil. However, MPE often fails to meet remediation targets for chlorobenzene–contaminated sites. To address this, researchers have developed a combined in–situ chemical oxidation (ISCO) and MPE synergistic remediation approach, which has demonstrated effective long–term stability in treating complex chlorobenzene groundwater pollution(Chen Yaojun, 2023).

Overall, Ex–situ remediation technology is suitable for large contamination areas and deeply buried contaminants. However, it requires transporting groundwater and surrounding soil, involves complex excavation and transportation work, and it may cause a certain degree of damage to the surrounding underground rock and soil environment during extraction. The excavated contaminated soil or groundwater requires secondary treatment to remove contaminants which makes the process complex and costly.

3.2. In–situ remediation technology for groundwater

In–situ remediation technology does not require changing the location of contaminated groundwater and remediates it at the contamination source. It offers advantages of low cost, reduced pollutant exposure, and minimal environmental disturbance. Compared to Ex–situ remediation technology, it requires less investment and causes less environmental interference. This makes this technology more promising for popularization (Chen Nanwei, 2019). Typical In–situ groundwater remediation technologies mainly include Groundwater circulation wells, Permeable reaction barriers, In–situ chemical oxidation, In–situ electrokinetic remediation, In–situ bioremediation, Nano–remediation, Monitored natural attenuation. Though seems good, this method contains some flaws and limitations such as lesser degradation potential of microbial community during on–site remediation, less bioavailability of pollutants on temporal and spatial scales, and lack of benchmark standards for efficiency and pollutant toxicity testing of bioremediation for their increased field applicability (Kuppusamy S, Palanisami T, Megharaj M, *et al.*, 2016).

3.2.1. Groundwater circulation wells (GCW)

Based on how the circulation is generated in circulation wells, this technology can be divided into Mechanical pump type, Air lift type and In–situ thermal treatment. Mechanical pump type circulation wells use mechanical pumps to extract wastewater and remove contaminants before reinjecting it underground to form a groundwater circulation. Air lift circulation wells inject air into groundwater through air pumps to form a water–vapor mixture. At this point, volatile contaminants in the groundwater transfer from the dissolved phase to the gaseous phase through rising bubbles. Due to the density difference between the water–vapor mixture and the outside environment, the mixture rises to the water–gas separation point. After the gas escapes, the contaminants are treated accordingly, and the water returns to the aquifer, forming a circulation. This technology is most effective in removing soluble volatiles in water. GCW technology does not require complex surface treatment engineering equipment and does not require extracting water to the surface, causing minimal disturbance to groundwater. The oxygen transfer process can promote the degradation of pollutants by aerobic organisms. It has the advantages of requiring few equipment, easy installation, and simple operation. In–situ thermal treatment technology is an improvement on air lift technology that heats contaminated groundwater, prompting contaminants and groundwater to convert to a gaseous state, which is then extracted to the surface for treatment using extraction wells. Compared to single circulation well technology, the remediation effect improved by 25.8% and effectively avoided tailing phenomena(Lu Liang, 2023).

3.2.2. Permeable reactive barrier (PRB)

PRB involves installing a permeable reactive barrier (PRB) composed of active materials along the groundwater flow path. As contaminated water passes through the barrier under natural hydraulic gradients, pollutants are removed or detoxified through processes including precipitation, adsorption, redox reactions, and biodegradation, thereby achieving groundwater purification. The most critical component of PRB technology is the reactive media filled within the wall. Its reactivity, stability, cost, and lifespan directly impact the treatment effect and economic cost of PRB for contaminated groundwater (Yang Rongrong, *et al.*,2021). PRB can degrade or immobilize contaminants *in situ* without bringing them up to the surface. Hence, there is no need to build above ground facilities other than monitoring wells as they are of high cost, and after the installation, the above ground can be re–used for variety of purposes (Thiruvenkatachari R, Vigneswaran S, Naidu R, 2008). However, Due to mineral precipitation, gas production, pH effects, and biofilm action, PRB may experience the problems of passivation and clogging issues. Therefore, analyzing the influencing factors and operating conditions for different passivation and clogging phenomena, improving PRB structural design, and enhancing operational efficiency and lifespan are important research directions for PRB in the future (Li Zhijian, Wei Li, Ni Heng, 2022).

3.2.3. In–situ chemical oxidation (ISCO)

The method involves direct injection of chemical oxidants into subsurface contamination zones through vertical or horizontal wells, where organic pollutants are degraded into harmless or low–toxicity byproducts via redox reactions, achieving groundwater remediation. Common oxidants used in this technology include Fenton's reagent, permanganate, O_3 and persulfate. Numerous researchers have conducted pioneering explorations regarding the application efficacy and operational requirements of chemical reagents. O_3 as an oxidant in ISCO technology to remediate groundwater contaminated with petroleum. The results showed that the highest degradation rates of benzene series and naphthalene in the contaminated groundwater reached 99.6% and 100% respectively, meeting the remediation targets(Nimmer *et al.*, 2000). However, in–situ permanganate oxidation resulting in decreased permeability, O_3 gas–phase mass transfer issues, and the strong acidity required for Fenton's reagent limit the development of these technologies. Inaccurate oxidant dosing can have negative effects, which has raised some doubts about the technology's development. Using the time required to reduce pollutant concentration to the target limit under oxidation as the measurement time for oxidant demand, while simultaneously using expressions with testing conditions to more accurately provide oxidant demand(Chen Kai, 2023). Deep mixing technology, proving that it is suitable for in–situ chemical oxidation remediation of organic–contaminated sites with deep and lightly contaminated areas. It has the advantages of mature technology, uniform mixing, small footprint, minimal environmental impact, high cost–effectiveness, and excellent remediation effect (Liu Zhiyang, 2023) .

3.2.4. In–situ electrokinetic remediation (EKR)

This technology is a relatively environmentally friendly remediation technology. Its operating principle involves inserting electrodes into contaminated zones and applying voltage to concentrate pollutants in electrode areas through ionic migration, electro–osmosis, and electrophoresis under electric field effects, followed by targeted removal from groundwater via physical or chemical methods. This method is mainly used to remove some organic pollutants and heavy metal ions. EKR operates without requiring additional chemical reagents and generates no adverse environmental impacts during the treatment process. It exhibits three core characteristics: Broad–spectrum contaminant compatibility, Precision–controlled directional pollutant migration, Zero secondary pollution. This method attracts increasing attention in recent years. Since the development and application of this technology depend on electrokinetic remediation devices, research on the remediation mechanism and processes of these devices is an important trend in this field.

3.2.5. In–situ bioremediation

This technology utilizes engineered interventions to stimulate or introduce specific microorganisms (indigenous or exogenous strains) within the native groundwater environment. Through their metabolic activities, these microbes degrade, transform, or immobilize contaminants in soil and groundwater, thereby achieving in situ reduction of both pollutant concentrations and toxicity. This technology is suitable for remediation after the underlying microbial flora system in the water body is destroyed, when the water body contains pollutants that are difficult for conventional microorganisms to degrade, or when specific microorganisms are difficult to become dominant flora (Gao Yesong, Wang Haihai, Wu Di, 2021). This technology has core advantages of low cost and minimal environmental disturbance. However, it is necessary to ensure that the microorganisms added are harmless to the environment, and there are high requirements for the operation procedures and product storage.

3.2.6. Nano–remediation technology

The material nano–mediation uses can effectively react with the pollutants because of its physical and chemical properties, like high specific surface area and reactivity. Additionally, the ecotoxicity of modified or coated nanomaterials can be mitigated to decrease its possible effect to the environment. This method can be separated into the following parts: nano zero–valent iron (nZVI) remediation technology, carbon nanotube (CNT) remediation technology, and metal and magnetic nanoparticle (MNP) remediation techniques (Alazaiza M Y D, Albahnasawi A, Ali G A M, *et al*., 2021). The nano–remediation is a remediation method in which nanomaterials could be included in presence of plants or biological agents. Therefore, scientists need to find out plant physiology, biochemistry and degradation of pollutants as well as a suitable assessment of probable collaborative impacts(El–Ramady H, El–Henawy A, Amer M *et al*., 2020).

Recent years, nZVI's preparation has been advanced quickly from the conventional chemical reduction method to biological and electrochemical methods. In that case, its stability largely increased. nZVI's usage has been extended: in the remediation of soil, groundwater, sediment, and other media contaminated by an increasing variety of pollutants; CNTs have special structures and properties like high specific surface area, strength, and conductivity. It can be used to remove heavy metals, organic pollutants, and bacteria, from water and soil; Metals and MNPS are widely used because of its special and unique absorption ability. The main advantage of it is that it is easy to be separated from magnetic, which makes it recyclable.

3.2.7. Monitored natural attenuation (MNA)

This technology utilizes natural processes to decompose and alter contaminants in groundwater. It relies on naturally occurring biodegradation, dispersion, sorption, dilution, volatilization, and other attenuation processes within contaminated sites to reduce the concentration, total quantity, and toxicity of pollutants in wastewater to acceptable risk levels within a reasonable timeframe. Therefore, it is also known as intrinsic bioremediation, natural recovery, or natural assimilation. This technology is characterized by low costs and the ability to effectively reduce exposure to pollutants and disturbance to the site environment. However, it requires simulation of the degradation rate and migration pathways of pollutants and prediction of pollutant concentrations at observation points with descending gradients when the contamination plume spreads. Researchers studied the natural attenuation characteristics of 1,2,3–trichloropropane (TCP) and benzene in different aquifers at a chemical contamination site through a three–year long–term monitoring of groundwater, and calculated the natural attenuation capacity of TCP and benzene in the pollution source area using a first–order decay kinetic equation (Li Hui *et al*., 2023).

Overall, single technology types can no longer meet the needs of treating various types of water sources with different contamination types and levels. The application of diversified remediation technologies for complex water source pollution will become the mainstream remediation method. For example, combined remediation technologies such as permeable reactive barriers (PRBs) and electrokinetic remediation, the integration of in–situ chemical oxidation (ISCO) with PRBs, multi–phase extraction combined with in–situ chemical oxidation, and electrokinetic–microbial remediation processes have been widely used in water pollution treatment.

4. Conclusion and outlook

4.1 The coupling of multiple technologies is an inevitable trend in future groundwater pollution treatment

The research on the analysis and remediation of groundwater pollution has evolved from the aplication of single methods and tools to the integrated use of multiple tools and coupling of various methods. The interdisciplinary infiltration, advancements in analytical testing methods, and the development of computer information technology have made pollution tracing more accurate and comprehensive. The comprehensive application of various remediation methods can achieve more efficient pollution remediation with lower costs and risks.

4.2 Big data technology will play a greater role in the remediation of groundwater pollution

It will become an important tool for analyzing groundwater pollution sources, assessing treatment effects, and predicting development trends. Driven by big data technology, combined with environmental isotope technology, microbial and water quality analysis technology, digital simulation and information technology, biological and composite remediation technology, and other technical methods, innovative approaches for groundwater pollution remediation with higher efficiency, lower costs, and reduced risks will surely be realized.

4.3 To protect groundwater, prevention is better than remediation

Although significant achievements have been made in tracing groundwater pollution sources and remediation, every method has its limitations, and even secondary pollution may be caused during the treatment process. Currently, no method can completely treat water pollution. The fundamental approach to protecting groundwater is to establish an environmental protection mindset and control and treat initially polluted water from the source. Rational selection of process flows, site layouts, wastewater recovery, and treatment is necessary to avoid falling into a cycle of pollution–treatment–re–pollution–re–treatment. Water resource protection and wastewater treatment are crucial projects for safeguarding green mountains and clear waters and creating a good ecological environment, which require the concerted efforts of the entire society.

Conflicts of interest

The author declares no conflict of interest.

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