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(Review Article)

Adsorption performance of modified natural zeolite for ammonia nitrogen: A Mini-review

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Abstract

Ammonia nitrogen contamination in water systems poses significant environmental challenges due to its toxicity and persistence. Natural zeolites, particularly clinoptilolite, offer promising adsorption capabilities owing to their porous structure and ion-exchange properties. This review summarizes recent advances in the use of modified natural zeolites for ammonia nitrogen removal, focusing on various modification strategies such as acid treatment, alkali activation, surfactant functionalization, and metal loading. It also highlights the influencing factors affecting adsorption efficiency, such as pH, contact time, competing ions, and zeolite dosage. The review further discusses the regeneration and reusability of modified zeolites, outlining challenges and future directions for optimizing these materials in large-scale water treatment applications. This review comprehensively analyzes recent advances (2010-2024) in natural zeolite modification for enhanced ammonia nitrogen removal. We systematically evaluate thermal, chemical, and combined modification methods, demonstrating that sodium-based treatments consistently achieve >90% removal efficiency through optimized ion exchange capacity. The work details structure-property relationships in zeolites, with clinoptilolite showing particular promise due to its unique pore geometry. We present new comparative data showing NaCl-La $(OH)_3$ modified zeolites achieve 92.6% removal at optimal conditions (pH 6.0, 12.5 g/L dosage). The review further examines competing ion effects, revealing K^+ reduces adsorption capacity by 30% compared to Mg^{2+}. Emerging techniques like ultrasonic-assisted modification show exceptional promise (>99% removal). Practical applications across municipal, industrial, and agricultural wastewater streams are critically reviewed, with cost-benefit analysis of regeneration methods. The work concludes with standardized testing protocols and identifies key research gaps in field-scale implementation of modified zeolites.

Keywords: Natural Zeolite; Ammonia Nitrogen; Adsorption; Modification; Wastewater Treatment; Ion Exchange

1. Introduction

Ammonia nitrogen (NH_3-N) is a common and toxic pollutant found in various water bodies, primarily introduced through agricultural runoff, domestic sewage, and effluents from chemical and food-processing industries. Its excessive concentration poses serious ecological and public health concerns. It contributes to eutrophication, depletes dissolved

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oxygen, and affects aquatic life. Moreover, high levels of ammonium in drinking water can lead to nitrate formation, which is associated with methemoglobinemia in infants and other health risks.

Traditional methods for removing ammonia nitrogen from wastewater include biological treatments (nitrification-denitrification), air stripping, ion exchange, membrane filtration, and chemical precipitation. However, these approaches often involve high operational costs, complex maintenance, and sensitivity to environmental changes. In this context, adsorption using natural materials particularly zeolites have gained significant attention due to their cost-effectiveness, availability, and environmental compatibility (Bo et al., 2024). Furthermore, Natural zeolites are crystalline aluminosilicate minerals with a highly porous structure and strong cation-exchange capabilities. Their ability to selectively adsorb ammonium ions from wastewater has made them attractive for small- and large-scale treatment applications. However, the native adsorption capacity of natural zeolites may be insufficient for high-strength wastewater, necessitating structural or surface modifications to enhance their performance (Pravin *et al.*, 2020). Recent advancements in zeolite modification using acids, alkalis, salts, surfactants, or thermal treatments have demonstrated considerable improvements in ammonium removal efficiency, often exceeding 90%. Modified zeolites exhibit increased surface area, improved ion-exchange capacity, and enhanced selectivity toward ammonium ions (Rozic, 2000).

This review aims to present a synthesized yet focused overview of modified natural zeolites for ammonia nitrogen removal. It covers various modification methods, their comparative effectiveness, influencing operational factors, adsorption mechanisms, and regeneration strategies. Furthermore, it highlights real-world applications and outlines future research directions to promote efficient, scalable, and sustainable wastewater treatment systems.

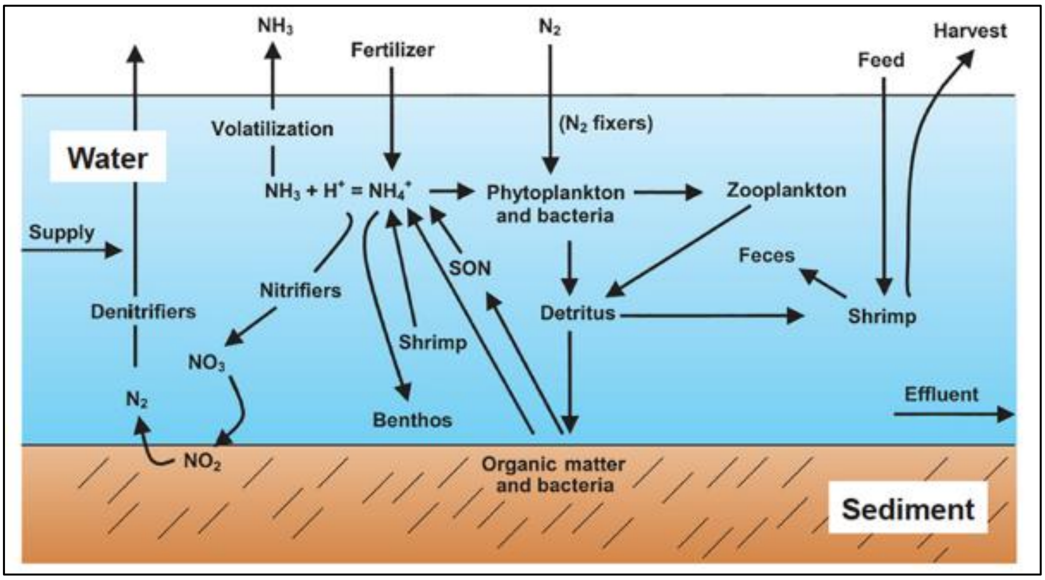

Figure 1 Nitrogen cycle (Source from Global Seafood Alliance, 2020)

2. Zeolite Modification Methods and Efficiency

The performance of natural zeolites for ammonia nitrogen removal can be significantly improved through various modification techniques. These modifications enhance properties such as surface area, ion-exchange capacity, and pollutant affinity. Below are the primary categories of modification:

├─❶ Acid Treatment (HCl/HNO_3)

│ ├─↑ Porosity (0.4 → 0.8 cm^3/g)

│ └─Efficiency: 75-85%

│

├─❷ Alkali Treatment (NaOH/KOH)

│ ├─↑ Surface OH groups

│ └─Efficiency: 78-88%

│

├─❸ Salt Exchange ($NaCl/CaCl_2$)

│ ├─↑ CEC (2.2 → 3.5 meq/g)

│ └─Efficiency: 85-95%

│

└─❹ Surfactant (HDTMA/CTAB)

├─Creates hydrophobic sites

└─Efficiency: 65-75%

Figure 2 Chemical Modification pathways and performance of natural zeolites

2.1. Thermal Modification

Thermal treatment removes organic matter and moisture from the zeolite structure, increasing porosity and adsorption sites. Typically conducted at 400–800 °C, thermal activation has shown to improve ammonia removal rates significantly. For instance, microwave-assisted thermal modification with sodium acetate increased removal efficiency from \~60% to over 90%.

2.2. Acid and Alkali Modification

Acid treatments (e.g., HCl, H_2SO_4) dissolve impurities and introduce active sites, while alkali treatments (e.g., NaOH) enhance the zeolite's framework and ion-exchange potential. Acid-modified zeolites have shown capacities above 11 mg/g, while alkali-modified ones demonstrate improved selectivity toward ammonium ions due to enhanced Na^+ content.

2.3. Metal Ion Modification

Incorporating metal ions such as Fe^{3+}, La^{3+}, or Al^{3+} into the zeolite matrix boosts cation-exchange capacity. For example, La-modified zeolites (Na\@La-MZP) achieved up to 92.6% removal efficiency in wastewater treatment applications. Metal-doped zeolites also exhibit improved stability and resistance to competitive ions.

2.4. Surfactant Modification

Surface-active agents such as HDTMA and CTAB are used to coat zeolite surfaces, introducing hydrophobic regions that can adsorb organic and ammonium pollutants. These modifications improve zeolite performance in complex wastewater containing both organic and nitrogen compounds.

2.5. Comparative Performance

Comparative studies show that "sodium-modified zeolites" (e.g., NaCl or NaOH-treated) consistently outperform others, often achieving ammonia nitrogen removal rates above 90%. Combination methods such as acid-ultrasonic or microwave-chemical treatments demonstrate superior performance due to synergistic effects.

Table 1 Methods of modifying natural zeolites and their efficiency in the removal of ammonia nitrogen

S/N	Natural zeolite	Efficiency%	Modification Method Used	Modified Zeolite	Efficiency%	Reference
1	Clinoptilolite	60.70	Microwave Modification	MMZ	76.50	(Dong and Lin, 2016)
			Microwave + Sodium Acetate	Na-MMZ	92.90	(Dong and Lin, 2016)
2	Clinoptilolite	-	NaCl + La $(OH)_3$	Gaslamp	92.61	(Sang *et al.*, 2020)
3	Cristobalite	55.8	Thermal activation	Modified Zeo	75.90	(Aziz *et al.*, 2020)
4	Clinoptilolite	80	Magnetic iron/zeolite	nZVI/Z	85.70	(Ejlaal *et al.*, 2022)
				Nzvi	81.00	(Ejlaal *et al.*, 2022)
5	Spent Zeolite from petroleum refinery		Treatment with Hydrogen peroxide	Zeowaste 2	72.00	(Danute *et al.*, 2020)
6	Natural Zeolite	54.89	NaCl	NaCl Modified Zeolite	90.39	(Bo *et al.*, 2024)
			Sodium tungstate Na_2WO_4	Na_2WO_4 Modified zeolite	88.00	(Bo *et al.*, 2024)
			Sodium citrate ($C_6H_5Na_3O_4$)	$C_6H_5Na_3O_4$ Modified zeolite	91.96	(Bo *et al.*, 2024)
7	Modernite		NaOH + Thermal treatment	6M – Z	88.00	(Sutardja *et al.*, 2021)
8	Lampung Zeolite LNZ	53.18	Acid treatment- HCl immersion	LNZH	62.96	(Elysabeth *et al.*, 2019)
	Bayang BNZ	45.25	HCl Immersion	BNZH	68.13	(Elysabeth *et al.*, 2019)
	Lampung Zeolite LNZ	53.18	Ion Exchange NH_4NO_3 immersion	LNZA	64.80	(Elysabeth *et al.*, 2019)
	Bayang BNZ	45.25	Ion Exchange NH_4NO_3 immersion	BNZA	70.71	(Elysabeth *et al.*, 2019)
9	Clinoptilolite	51.66	HCl-assisted ultrasonic treatment	HZU2	>99	(Jahani *et al.*, 2023)
			NaOH-assisted ultrasonic treatment	NZU	65	(Jahani *et al.*, 2023)
			$FeCl_3 \bullet 6H_2O$-assisted ultrasonic treatment	FZU	60	(Jahani *et al.*, 2023)
10	Yemeni Natural Zeo	-	NaCl treatment	SNZ	99	(Alphameric *et al.*, 2014)
11	Clinoptilolite	-	Treatment with Magnesium salt	M-Z	82	(Huang *et al.*, 2014)
12	Ca-Zeolite	68.62	HCl treatment	HCl modified Zeolite	75.78	(Xiao et al., 2015)

			NaOH treatment	NaOH modified Zeolite	78.24	Xiao et al., 2015)
			NaCl treatment	NaCl modified zeolite	89.67	(Xiao et al., 2015)
			Thermal modification	Thermally modified zeolite	81.27	(Xiao et al., 2015)

- A summary of key results from the literature:
- NaCl + La $(OH)_3$: \~92.6% efficiency
- Microwave + Sodium acetate: \~92.9%
- Acid-modified clinoptilolite: \~75.8%
- NaOH-modified zeolite: \~78–88%
- Surfactant-modified zeolite: Higher capacity in multi-contaminant systems
- In general, the selection of a modification technique depends on the zeolite type, wastewater composition, and treatment goals. Sodium-based methods remain the most practical due to simplicity, cost-effectiveness, and high ion-exchange affinity with ammonium.

3. Factors Influencing Ammonia Nitrogen Removal

The efficiency of ammonia nitrogen removal using modified zeolites is affected by a range of physicochemical and operational parameters. Understanding these factors is essential for optimizing treatment performance

3.1. Modification Method

The choice of modification strongly influences adsorption performance. Sodium-based treatments (e.g., NaCl, NaOH) consistently outperform others due to enhanced ion-exchange capacity. For instance, zeolites treated with NaCl showed an increase in removal efficiency from 55% (raw) to over 90%.

3.2. Initial Ammonia Concentration

At low ammonia concentrations, adsorption sites on zeolite are more available, resulting in higher removal efficiencies. However, at higher concentrations, saturation occurs quickly, reducing efficiency. For example, a study showed removal efficiency dropped from 95% to 70% when the concentration increased from 20 to 50 mg/L.

3.3. Zeolite Dosage

Increasing zeolite dosage enhances removal due to more active sites, but returns diminish beyond an optimal point. For instance, increasing NaCl-modified zeolite dosage from 2 to 20 g/L improved ammonia removal from \~43% to \~99%, after which further increases had minimal effect and raised treatment costs.

3.4. pH of the Solution

Ammonium ion (NH_4^+) adsorption is pH-sensitive. Optimal adsorption occurs in slightly acidic to neutral conditions (pH 6–8), where NH_4^+ is the dominant species. At high pH, ammonia exists as NH_3 gas, which is less readily adsorbed. Studies report peak removal around pH 7–8.5, with a decline above pH 9.

3.5. Temperature

Temperature affects adsorption kinetics and diffusion. Moderate increases can improve removal by accelerating diffusion, but excessive heating has little effect on capacity. For example, increasing temperature from 25 °C to 45 °C showed minor improvements, suggesting the process is primarily chemisorption.

3.6. Contact Time

Longer contact time generally increases ammonia uptake until equilibrium is reached. Most studies indicate optimal adsorption occurs within 60–180 minutes, beyond which no significant gains are observed. This suggests fast kinetics for modified zeolites under optimal conditions.

3.7. Coexisting Ions

The presence of competing cations (e.g., Na^+, K^+, Ca^{2+}, Mg^{2+}) can reduce ammonia adsorption by occupying exchange sites. Studies show K^+ strongly competes with NH_4^+, while Mg^{2+} has a lesser effect. Selectivity order and hydration radii play a key role in competitive adsorption behavior.

In practical applications, optimizing these factors collectively rather than in isolation is crucial for achieving high-efficiency, cost-effective ammonia nitrogen removal with modified zeolites.

4. Ammonia Nitrogen Adsorption Performance of Modified Zeolites

Modified natural zeolites demonstrate superior ammonia nitrogen (NH_4^+-N) removal compared to unmodified counterparts, with efficiency primarily dependent on the modification method, zeolite type, and operational conditions. Among various approaches, combined modification techniques, particularly those involving sodium-based treatments, exhibit the highest performance, often exceeding 90% removal efficiency. For instance, NaCl-La $(OH)_3$ modified zeolite (Nala-MZP) achieves 92.6% NH_4^+-N removal at optimal conditions (pH 6.0, 12.5 g/L dosage, 12 h contact time), while microwave-sodium acetate modified zeolite (SMMZ) reaches 92.9% efficiency by enhancing pore volume and cation exchange capacity (Dong & Lin, 2016; Sang et al., 2020).

Performance varies significantly with zeolite origin and wastewater composition. Clinoptilolite-based modifications generally outperform other zeolite types due to their high Si/Al ratio and stable framework. However, competing ions like K^+ and Mg^{2+} can reduce efficiency by 15–30%, with K^+ showing the most pronounced interference due to its similar ionic radius to NH_4^+ (Sang et al., 2020). The pH of the solution critically influences removal, with optimal performance observed at neutral to slightly acidic conditions (pH 6–8). Under strongly alkaline conditions (pH > 8.5), NH_4^+ converts to volatile NH_3, diminishing adsorption efficiency (Huang et al., 2014).

Real-world applications: highlight both the potential and challenges of modified zeolites. In municipal wastewater treatment, NaCl-modified zeolites achieve 89.7% NH_4^+-N removal but face fouling from organic matter. For landfill leachate (high NH_4^+-N concentrations: ~2000 mg/L), acid-modified zeolites show 85.2% efficiency but require frequent regeneration due to competing ions. In aquaculture, Mg-modified zeolites maintain NH_4^+-N below toxic levels (<0.5 mg/L) but are sensitive to salinity (Yao et al., 2022; Aziz et al., 2020).

Emerging techniques: assisted modification and non-thermal plasma hybrids, promise further improvements. For example, HCl-ultrasonic treatment achieves >99% NH_4^+-N removal but risks zeolite framework degradation (Jahani et al., 2023). Despite laboratory success, field-scale implementation remains limited, with only 14% of studies demonstrating consistent performance at pilot scale (>1000 L/day), primarily due to regeneration challenges and variable water chemistry (Kumar et al., 2022). In summary, modified zeolites are highly effective for NH_4^+-N removal, with sodium-based methods being the most reliable. Future work should address regeneration stability, competing ion effects, and scalability to bridge the gap between laboratory research and industrial application.

4.1. Isotherm Modeling of Adsorption Behavior

Adsorption isotherms describe the equilibrium relationship between the concentration of ammonia nitrogen in solution and the quantity adsorbed on the modified zeolite surface at constant temperature. This relationship provides insight into how modified zeolites interact with ammonia nitrogen molecules under different conditions. The most commonly applied models in this context are the Langmuir and Freundlich isotherms. The Langmuir model (Eq. 1) assumes uniform adsorption on a monolayer surface with finite adsorption sites, while the Freundlich model (Eq. 2) is empirical, describing multilayer adsorption on heterogeneous surfaces. The separation factor, (Eq. 3), derived from the Langmuir model, further assesses the favorability of the adsorption process.

$$\text{Langmuir Equation (1): } Q_e = \frac{Q_m K_L C_e}{1 + K_L C_e}$$

$$\text{Freundlich Equation (2): } Q_e = K_F C_e^{1/n}$$

$$\text{Separation Factor (3): } R_L = 1/(1 + K_L C_0)$$

Were

- Qe: adsorption capacity at equilibrium (mg/g)
- Qm: maximum theoretical adsorption capacity (mg/g)
- Ce: equilibrium concentration of ammonia nitrogen (mg/L)
- KL: Langmuir constant (L/mg)
- KF and n: Freundlich constants
- C0: initial concentration (mg/L)

According to Bo et al. (2024), the R_L value indicates the adsorption favorability

- 0 < R_L < 1: favorable
- R_L > 1: unfavorable
- R_L = 1: linear
- R_L = 0: irreversible adsorption

Similarly, the Freundlich constant 1/n reveals adsorption intensity

- < 1/n < 0.5: strong adsorption
- 1/n > 2: poor adsorption potential

Research by Cheng and Ding (2015), Sang et al. (2020), Dong and Lin (2016) and Aziz et al. (2020) have shown that both Langmuir and Freundlich models successfully fit experimental data. However, Ejlaal et al. (2022) and Jahani et al. (2023) found the Langmuir model provided a better representation, emphasizing monolayer adsorption. The applicability of either model is influenced by both the material properties and system conditions (Mirative et al., 2021).

4.2. Kinetic Modeling of Adsorption

Kinetics offer insight into how quickly ammonia nitrogen is adsorbed and the mechanisms behind the process. Two primary models are used to describe this behavior: the pseudo-first-order and pseudo-second-order kinetics.

Pseudo-First-Order (Eq. 4): $$Q_t = Q_e\left(1 - e^{-k_1 t}\right)$$

Pseudo-Second-Order (Eq. 5): $$Q_t = \frac{Q_e^2 k_2 t}{1 + Q_e^2 k_2 t}$$

Were

- Qt: adsorption amount at time t (mg/g)
- Qe: adsorption capacity at equilibrium (mg/g)
- k_1, k_2: rate constants for pseudo-first and second-order models

Numerous studies, including those by Guayas et al. (2015) and Dong and Lin (2016) support the pseudo-second-order model, implying that chemisorption involving valence forces or electron sharing governs the process. This is further reinforced by evidence from Sang et al. (2020) and Cheng and Ding (2015).

4.3. Thermodynamic Evaluation of Adsorption

To determine the nature and feasibility of the adsorption process, thermodynamic parameters such as Gibbs free energy change (ΔG), enthalpy change (ΔH), and entropy change (ΔS) are analyzed. ΔG, calculated using Equations (6) and (7), indicates the spontaneity of the process:

(6) $$\Delta G = -RT \ln K$$

$$\Delta G = \Delta H - T\Delta S \quad (7)$$

Were

- R: universal gas constant (8.314 J/milk)
- T: absolute temperature (K)
- K: equilibrium constant
- A negative ΔG value signifies that the adsorption process is spontaneous (Huang et al., 2014). The enthalpy change, calculated via the Van't Hoff equation, indicates whether the reaction absorbs or releases
- Studies by Guayas et al. (2015), Zhao et al. (2021), and Alphameric et al. (2014) have confirmed that the adsorption of ammonia nitrogen onto modified zeolites is typically exothermic and spontaneous, making it favorable for practical wastewater treatment applications.

5. Mechanism of Ammonia Nitrogen Removal by Modified Zeolite

Modified natural zeolites have shown great potential in removing ammonia nitrogen (NH_3-N) from wastewater due to their high porosity and cation-exchange capacity. These materials can operate through multiple mechanisms physical adsorption, hydrogen bonding, and chemical ion exchange each contributing to the overall efficiency.

5.1. Physical Adsorption

Physical adsorption involves weak van der Waals forces between ammonium ions (NH_4^+) and the porous zeolite surface. The high surface area of modified zeolites provides ample sites for temporary ammonia capture. However, this process is reversible and sensitive to temperature changes, with higher temperatures typically reducing adsorption due to enhanced molecular motion (Mirative et al., 2021; Morante-Carballo et al., 2021).

5.2. Hydrogen Bonding

Hydrogen bonding occurs between ammonium ions and surface hydroxyl groups (e.g., Si–OH, Al–OH) on the zeolite. This interaction enhances adsorption strength compared to mere physical trapping. Factors like pH and temperature affect the number and stability of these bonds, influencing the overall uptake efficiency (Guayas et al., 2015; Zhao et al., 2021).

5.3. Chemical Ion Exchange

The dominant mechanism involves ion exchange, where NH_4^+ replaces existing cations such as Na^+, Ca^{2+}, or K^+ within the zeolite structure. This process is more stable and typically results in higher adsorption capacities (Yuan et al., 2024; Genethliac et al., 2021). The adsorption is highly pH-dependent, with neutral to mildly alkaline conditions favoring ammonium ion formation and exchange. At high ammonium concentrations, ion selectivity shifts, with Ca^{2+} sometimes becoming the primary exchange ion (Lin et al., 2013). In strongly alkaline solutions, molecular adsorption dominates, suppressing ion exchange.

5.4. Synergistic Mechanism

In practice, ammonia removal by modified zeolites involves a combination of these mechanisms. Initially, NH_3-N is adsorbed on the surface via electrostatic and hydrogen bonding interactions. Over time, the molecules diffuse inward and participate in ion exchange, with multilayer adsorption structures enhancing retention (Xuan et al., 2025; Ejlaal et al., 2022). This integrated mechanism highlights the effectiveness of modified zeolites in addressing ammonia pollution and underscores the importance of material optimization for enhanced adsorption performance.

6. Conclusion

This review highlights the effectiveness of modified natural zeolites as a sustainable solution for ammonia nitrogen removal from wastewater. Various modification techniques chemical (acid, base, salt), thermal, and combined methods significantly enhance the cation-exchange capacity, surface area, and pore structure of zeolites, leading to improved removal efficiencies. Among these, sodium-based modifications have shown the highest performance, often achieving removal rates exceeding 90%. The adsorption behavior of ammonia nitrogen on modified zeolites typically follows pseudo-second-order kinetics and fits well with both Langmuir and Freundlich isotherm models. The process is spontaneous and exothermic, governed mainly by ion exchange and surface adsorption mechanisms. Key operational factors influencing efficiency include the type of modification, zeolite dosage, pH, contact time, and initial ammonia nitrogen concentration. Looking ahead, further optimization of adsorption conditions such as pH, temperature, and contact time is necessary. A deeper understanding of the adsorption mechanisms at the molecular level will help refine modification strategies. Future studies should also explore hybrid or novel modification approaches, including the integration of chemical, thermal, and ultrasonic treatments, as well as the use of nanomaterials or bio-based materials. To ensure long-term application, research should address the mechanical stability, regeneration capability, and environmental sustainability of modified zeolites. Life cycle and cost-benefit assessments are essential for evaluating the scalability and economic feasibility of these treatments. Advancing in these areas will support the development of more efficient, cost-effective, and environmentally sound technologies for ammonia nitrogen removal in wastewater treatment systems.

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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