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MAPPING THE POTENTIAL OF CHITOSAN IN REMOVING ANTIBIOTICS FROM AQUACULTURE WASTEWATER

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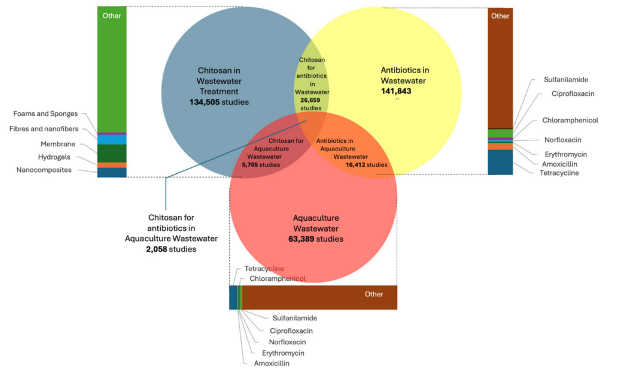
HIGHLIGHTS

- Chitin and chitosan are natural biopolymers with versatile properties for wastewater treatment.
- Alternative chitosan-based approaches can be used to address challenges in treating aquaculture wastewater.
- Chitosan hydrogels are highly effective for dye removal.
- Chitosan-based adsorbents may remove antibiotics like tetracycline.

GRAPHICAL ABSTRACT



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ABSTRACT

The potential of chitosan-based materials for the remediation of aquaculture wastewater is evaluated, emphasising emerging pollutants and the mechanisms underlying their adsorption processes. The efficiency of chitosan nanocomposites and their modifications in adsorbing antibiotics such as tetracycline is scrutinised, providing insights into deprotonation, protonation and the impact of concentration on surface interactions. Chemical modifications enhancing adsorption efficiency and the synergistic removal of antibiotics and metal ions using advanced materials like magnetic core-brush composites and cross-linked electro-spun chitosan nanofibers are highlighted. The discourse extends to the challenges and recent advancements in removing a spectrum of antibiotics, including tetracycline, amoxicillin, erythromycin, norfloxacin, chloramphenicol, ciprofloxacin, and sulphanilamide. Various adsorbents such as chitosan nanocomposites, hydrogels, membranes, fibres and nanofibers, and foam and sponges are examined alongside molecularly imprinted chitosan for selective adsorption. The optimisation of adsorption with chitosan-metal microspheres and the pivotal role of pH-dependent mechanisms and chemisorptive processes are also explored. In summary, chitosan-based materials demonstrate substantial promise for the efficient removal of antibiotics from aquaculture wastewater, with ongoing research dedicated to optimising adsorption capacities.

Introduction

Aquaculture has become an integral part of the global food supply chain, providing a substantial source of seafood and other aquatic products. However, its rapid expansion has also led to increased concerns about the environmental impact, particularly due to the discharge of wastewater containing diverse pollutants. These pollutants, termed as "emerging contaminants" include pharmaceutical residues, antibiotics, heavy metals, and microplastics, which pose significant risks to aquatic ecosystems and human health (Mishra *et al*., 2023). Aquaculture wastewater encompasses a complex mixture of organic matter, nutrients (nitrogen and phosphorus), suspended solids, pathogens, and chemicals. The composition of aquaculture effluents varies depending on the species being cultured, feed input and management practices. For instance, the water from finfish farms contains more nitrogenous compounds from uneaten feed and fish excretion, whereas shrimp pond water has a higher content of organic matter and antimicrobial agents due to the shrimps' lack of acquired immune system, making them vulnerable to pathogens and unresponsive to vaccination (Thornber *et al.*, 2020). The discharge of untreated or inadequately treated effluents can result in eutrophication, harmful algal blooms, oxygen depletion in waterways, and accumulation of persistent contaminants in sediments. These environmental impacts not only threaten aquatic biodiversity in the wild, but also eventually compromise the sustainability of aquaculture operations (Ahmad *et al*., 2021).

Emerging pollutants in aquaculture wastewater are of particular concern due to their potential to disrupt aquatic ecosystems and enter the food chain. Pharmaceutical residues like hormones and antibiotics, for example, can exert sub-lethal effects on aquatic organisms and contribute to the development of antimicrobial resistance. Microplastics can be ingested by marine life, leading to physical harm and bioaccumulation of toxic substances. The removal of emerging pollutants from aquaculture wastewater is thus critical to safeguarding environmental quality and human health, which has been the focus of more than 2,000 studies (Figure 1). Effective treatment technologies capable of selectively removing these contaminants are essential to mitigate their adverse impacts on aquatic ecosystems and maintain the sustainability of aquaculture practices.

Chitin, a natural biopolymer found in the exoskeletons of crustaceans and insects, its derivative chitosan have gained attention as promising materials for wastewater treatment due to their biocompatibility, biodegradability, and versatile functional properties. This review aims to provide a brief overview of chitin and chitosan-based modifications for the removal of emerging pollutants from aquaculture wastewater. Moreover, this review encompasses a comprehensive exploration of chitin and chitosan, focusing on their structural and functional properties relevant to wastewater treatment applications. It includes a detailed discussion on the diverse modifications and formulations of these materials designed to enhance the efficiency of pollutant removal. The review evaluates the potentials of these materials in aquaculture wastewater treatment, addressing associated challenges and limitations. By synthesising current knowledge and recent advancements in chitin and chitosan-based technologies, this review seeks to contribute to the understanding of their role in mitigating the environmental impacts of aquaculture wastewater and advancing towards more sustainable aquatic food production systems.

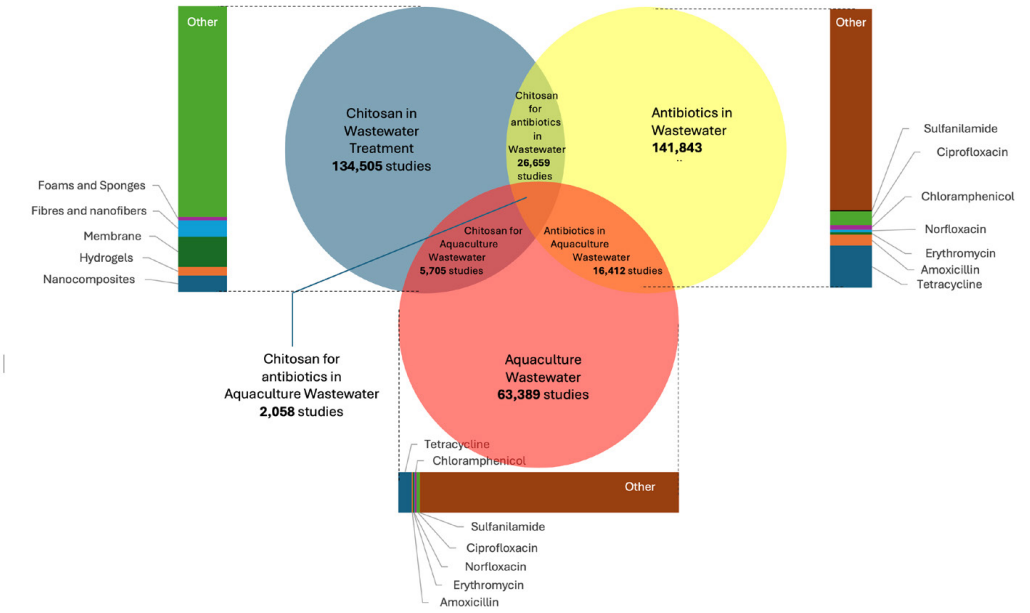

Figure 1: Number of studies performed on aquaculture wastewater (red), for antibiotic removal (yellow), and using chitosan treatment (blue). Data was extracted from Scopus using the keywords "Wastewater", "Antibiotics", "Aquaculture", and "Chitosan"

Modified Chitosan for Wastewater Treatment

Nanocomposites

The integration of nanomaterials has significantly transformed wastewater treatment research, introducing unprecedented efficiency. These nanomaterials exhibited exceptional performance largely due to their increased specific surface area, which is the total surface area per unit mass and is pivotal in defining the properties of solids. A higher specific surface area may enhance the "active sites" available for contaminant removal in wastewater. Furthermore, the quantum size effect, which alters the electronic properties of nanomaterials at the nanoscale is a crucial factor. While reducing particle size from macro to micro scale may not markedly affect electronic properties, transitioning to the nanometre scale can induce significant changes (Shukla *et al.*, 2013). Among the various nanomaterials, titanium dioxide (TiO_2) stands out in wastewater treatment applications. TiO_2 is highly favoured due to its affordability, robust chemical stability, wide availability, non-toxicity, and eco-friendliness. However, pristine TiO_2 is limited to absorbing light in the ultraviolet spectrum because of its large band gap of 3.2 eV. To overcome this limitation, extensive research has focused on band gap modification to enable TiO_2 activation under visible light (Leong *et al.*, 2015).

While nanoparticles show great potential in wastewater treatment, using them alone often is not enough. TiO_2, for example, is a highly effective photocatalyst that can break down many organic pollutants. However, it is not effective against non-biodegradable metal ions, inorganic compounds, and heavy metals (Chen & Ray, 2001). Additionally, TiO_2's indefinite lifespan means the nanoparticles itself may accumulate to toxic levels in food chains. Using suspended photocatalytic particles also introduces challenges like the need for a final recovery step (Szabó *et al.*, 2013) and the risk of nanoparticles being released into the environment, posing

hazards to humans, animals, and nature (Reijnders, 2009). The potential toxicity and complex regeneration process of photocatalysts may limit their industrial use (Gaya & Abdullah, 2008). To address these issues, researchers have been working since the 1980s to immobilise nanoparticles like TiO_2. They have explored various supports and binders to enhance the photocatalytic process while reducing the risk of pollution (Ali *et al.*, 2023). The use of chitosan as a binder for TiO_2has proven particularly effective. The amine and hydroxyl groups in chitosan provide binding and reaction sites for materials through adsorption, offering multiple ways to treat different wastewater pollutants (Ali *et al.*, 2023). This approach not only stabilises TiO_2, but also improves the overall efficiency and safety of the treatment process, making it more practical and sustainable for wastewater management.

Addressing the challenges associated with TiO_2, researchers have explored alternative approaches using chitosan-based materials for environmental applications. For instance, Hamdi *et al.* (2015) pioneered a hybrid photocatalyst by combining chitosan with phthalocyanine-TiO_2 (PC/CS-TiO_2), focusing on stabilising Phthalocyanine (PC) nanoparticles to prevent aggregation and ensure effective use. Their study emphasised the importance of chemical stability in optimising the nanocomposite performance across different pH levels for pollutant capture and degradation. Le Cunff *et al.* (2015) investigated chitosan-TiO_2's effectiveness in removing terbuthylazine (TBA), noting challenges such as potential titanium leakage under acidic conditions due to chitosan's pH-dependent solubility. Despite these hurdles, the nanocomposite demonstrated robust photocatalytic activity under UV light, effectively transforming TBA into non-toxic cyanuric acid after pretreatment with mildly acidic water (Le Cunff *et al.*, 2015). Jaiswal *et al.* (2012) expanded on these findings, demonstrating that copper-chitosan nanocomposites efficiently absorb organophosphate pesticides like malathion, particularly in mildly acidic water. They highlighted the nanocomposite's versatility across various pesticide concentrations, effectively transforming pollutants into harmless compounds, thus, demonstrating its potential for environmental clean-up. Additionally, Rezaei *et al.* (2019) utilised nano-chitosan derived from shrimp shells and activated carbon from date palm seeds to successfully remove nitrite, phosphate, and ammonia from aquaculture wastewater. Their optimisation of adsorption conditions, including pH levels, initial pollutant concentrations and contact times, resulted in high removal efficiencies, underscoring the efficacy of chitosan-based nanocomposites in water purification.

Chitosan-based nanocomposites offer diverse strategies for Cr (VI) removal, including polymer-grafted chitosan (Igberase *et al.*, 2014; Sethy & Sahoo, 2019), chitosan-clay nanocomposites (Li & Bowman, 2001; Gu *et al.*, 2019), magnetic chitosan-metal nanocomposites (Zhang *et al.*, 2016), and carbon-based chitosan nanocomposites (Chatterjee *et al.*, 2010; Beheshti *et al.*, 2016), each leveraging unique materials and methods to enhance adsorption efficiency and sustainability in water treatment. Chitosan-based nanocomposites have emerged as versatile materials for removing Cr (VI) from water due to their chemical modification capabilities (Sharma *et al.*, 2017). Polymer-grafted chitosan such as methyl methacrylate (MMA) grafted onto silica gel-crosslinked chitosan, exhibits enhanced mechanical strength but may experience reduced adsorption capacity at low pH levels due to competition from hydrogen ions with metal ions on the adsorbent surface (Igberase *et al.*, 2014; Sethy & Sahoo, 2019). Moreover, chitosan-polyacrylic acid nanocomposite hydrogels synthesised through chemical crosslinking have shown promise in removing chromium (VI) ions from aqueous solutions, following non-linear Redlich-Peterson isotherms and demonstrating monolayer formation and multisite interactions (Vilela *et al*., 2019).

Additionally, chitosan-crosslinked-poly (alginic acid) nanohydrogels have demonstrated high effectiveness in Cr (VI) removal, displaying spontaneous and chemical adsorption behaviour governed by pseudo-second-order kinetics (Sharma *et al*., 2017). Clay minerals, known for their unique structural, rheological and thermal properties have gained attention in environmental remediation, particularly for their potential in removing Cr (VI) from aqueous solutions (Li & Bowman, 2001; Gu *et al.*, 2019). Modified clays, like montmorillonite (MMT) and bentonite, when combined with chitosan through in situ graft polymerisation may form nanocomposites with enhanced adsorption capacities for Cr (VI) (Chang & Juang, 2004; Zhang *et al*., 2007). These nanocomposites utilise the unique bilayer adaptation of chitosan within the clay's interlayer space, demonstrating efficient removal mechanisms including chemical bonding and ion exchange (Darder *et al.*, 2003; Kumar *et al*., 2012). Chitosan-metal nanocomposites, particularly those incorporating magnetic properties are efficient adsorbents for Cr (VI) removal due to their biocompatibility and large surface area (Ambashta & Sillanpää, 2010). Magnetic chitosan nanocomposites can be easily separated using a magnetic field, simplifying purification and enabling reuse in water treatment applications (Zhang *et al*., 2016). Iron (Fe)-based chitosan composites, for example, effectively reduce Cr (VI) to less toxic Cr(III) states, utilising chitosan's hydroxyl (-OH) groups as electron donors (Shen *et al.*, 2013; Zhu *et al*., 2016; Cai *et al*., 2019). Nanocomposites like Fe_3O_4/SiO_2/CS-TETA possess hollow structures and modified surface functionalities, ensuring high adsorption capacities and rapid equilibrium times for Cr (VI) removal (Wang *et al*., 2020). Carbon-based chitosan nanocomposites, incorporating materials like multiwall carbon nanotubes and graphene oxide, offer high surface area and stability under extreme conditions for Cr (VI) removal (Chatterjee *et al*., 2010; Beheshti *et al*., 2016). Multiwall carbon nanotubes modified with chitosan also exhibit impressive adsorption capacities, overcoming challenges such as poor dispersion through acid oxidation and enhancing suitability for wastewater treatment (Vuković *et al*., 2010; Huang *et al*., 2018). Graphene oxide-chitosan nanocomposites, on the other hand, can leverage graphene oxide's hydroxyl and carboxyl (-COOH) groups for effective Cr (VI) adsorption, besides facilitating magnetic separation techniques for efficient pollutant removal (Ren *et al*., 2013; Samuel *et al*., 2018).

Hydrogels

Recent investigations have concentrated on the advancement of chitosan-based hydrogels, which demonstrate exceptional properties ideal for wastewater treatment applications. These hydrogels are synthesised through the formation of a three-dimensional network that allows for substantial swelling in water, thereby significantly increasing their adsorption capacity. The adaptability of chitosan hydrogels is attributed to their potential for various physical and chemical modifications aimed at enhancing their mechanical strength and adsorption efficiency. Physically, chitosan hydrogels can be altered through processes such as crosslinking, grafting and blending with other materials. Common crosslinking agents, including glutaraldehyde and epichlorohydrin, are employed to improve the mechanical properties of chitosan hydrogels, thereby increasing their durability for practical use (Pakdel & Peighambardoust, 2018). These agents covalently bond with chitosan, leading to a stable hydrogel structure. Nevertheless, the use of certain crosslinking agents can introduce toxicity, necessitating a careful balance between achieving mechanical robustness and maintaining biocompatibility.

Chemical modifications of chitosan hydrogels enhance their capacity for contaminant removal by grafting functional groups that target specific pollutants. For example, grafting with polyacrylic acid (Zhang *et al*., 2019) or polyacrylamide (Singh *et al*., 2009) increases the hydrogel's affinity for heavy metals and dyes by providing more active sites for contaminant adsorption. The inclusion of nanoparticles like graphene oxide (Medina *et al*., 2016) and metal

oxides (Priyadarshi *et al.*, 2022) further boosts adsorption efficiency due to the expanded surface area and improved interaction with pollutants.

The adsorption mechanisms of chitosan-based hydrogels are divided into chemisorption and physisorption. Chemisorption, involving the formation of chemical bonds is irreversible whereas physisorption, which depends on weaker physical forces such as van der Waals interactions, hydrogen bonding and hydrophobic interactions is reversible. The efficiency of adsorption is influenced by factors such as pH, ionic strength, adsorbent dose, initial pollutant concentration, contact time, and temperature (Ahmad *et al.*, 2015). Optimising these parameters is essential for designing effective industrial-scale adsorption processes. In a notable study, Li *et al.* (2024) developed a chitosan hydrogel with high adsorption performance for palladium (Pd) ions in industrial wastewater. This hydrogel, prepared using chitosan and acrolein as a cross-linking agent, demonstrated optimal Pd ion removal at pH 4, with an adsorption capacity of 505.05 mg/g under ideal conditions. The adsorption followed the pseudo-second-order kinetic model and the Langmuir model, with FT-IR and XPS analyses showing that the adsorption involved chelation of palladium(II) with amino ($–NH_2$) groups and electrostatic adsorption of $PdCl_{42}$- and $–NH_3$+ (Li *et al.*, 2024). This study underscores the potential of chitosan hydrogel adsorbents for commercial applications in treating industrial wastewater containing Pd.

Chitosan hydrogels are highly effective as dye adsorbents due to their low energy consumption, rapid and simple operation, considerable flexibility, biodegradability, and high adsorption rates (Liu *et al.*, 2018; Lu *et al*., 2018). Han *et al.* (2021) developed CMCS-PA composite hydrogels through the co-assembly and robust cross-linking interactions of carboxymethyl chitosan (CMCS) and phytic acid (PA). These hydrogels demonstrated excellent adsorption capacities for dyes such as Methyl Orange (MO) and Congo Red (CR), fitting pseudo-first-order and pseudo-second-order adsorption models. Additionally, these hydrogels exhibited significant adsorption reusability, stability and swelling ability, presenting a novel strategy for designing composite hydrogel materials for dye contamination control (Han *et al.*, 2021). The significant swelling capacity of chitosan facilitates the movement of water and dye molecules, enhancing the interaction between the hydrogel's binding sites and the dyes, thus, improving adsorption performance. The abundance of hydroxyl and amine functional groups in chitosan molecules enables the physical adsorption of food dyes (de Farias *et al*., 2018). In acidic aqueous solutions, chitosan acts similarly to cationic polymers, effectively interacting with anionic contaminants in wastewater, and thereby enhancing the removal efficiency of food dyes (de Farias *et al.*, 2018).

Le *et al.* (2018) synthesised chitosan hydrogels by activating chitosan with carbon dioxide (CO_2) and tested their adsorption and removal efficiency of vivid blue FCF and Congo red food dyes at various temperatures. The activation process resulted in the formation of three functional groups (protonated amino group, carbamate, and bicarbonate), where the NH_2 group serves as a site for dye adsorption and cross-linking. The activation of CO_2 in the chitosan network forms carbamate cross-links and the activation energy of the carbohydrates created by amine and CO_2 decreases at elevated temperatures, increasing the stability of chitosan in CO_2 treatment systems. The hydrogel's effectiveness in dye removal improves as the temperature rises, since CO_2 solubility or aqueous pH does not affect the stability of chitosan at higher temperatures. Additionally, Zhang *et al.* (2023) prepared a dual network hydrogel (CMCS/PVA) using carboxymethyl chitosan (CMCS) and polyvinyl alcohol (PVA) through freeze-thawing and cross-linking with calcium chloride. During preparation, –OH in PVA and –COOH in CMCS were physically crosslinked via hydrogen bonding, resulting in a hydrogel with a three-dimensional network structure. The esterification of the PVA ester

bond with CMCS further strengthened the link between CMCS and PVA. The carboxyl anion in carboxymethyl chitosan then interacted with the positively charged calcium ions in calcium chloride, leading to the formation of a hydrogel with a dual network structure. This hydrogel exhibited an increased pore structure, providing a larger surface area for adsorption. Functional groups like $–NH_2$, –COOH, and –OH in the hydrogel served as adsorption sites, enhancing the hydrogel's capacity to bind to AB dyes through electrostatic forces, hydrogen bonding and van der Waals forces.

A critical evaluation of the existing literature reveals that while significant progress has been made in enhancing the properties and performance of chitosan-based hydrogels, challenges remain. The synthesis of non-toxic, environmentally friendly crosslinking agents and the scalability of these hydrogels for industrial applications are areas that require further research. Moreover, the regeneration and reuse of chitosan hydrogels without significant loss of adsorption capacity are critical for sustainable wastewater treatment processes.

Membranes

Chitosan adsorbent membranes are characterised by their flat, thin structure, forming a continuous layer of chitosan or chitosan derivatives. Various methods can be employed to prepare these membranes. The casting method involves pouring a solution of chitosan dissolved in an organic acid such as acetic acid, onto a flat support such as a glass or polymer plate. The solution is evenly spread using a spatula or coating device, followed by air drying or controlled temperature drying to allow solvent evaporation and membrane formation (Nakayama *et al.*, 2020). Another method involves precipitation, where the chitosan solution is added to a precipitation bath containing distilled water or a mixture of water and alcohol such as methanol or isopropanol. The water/alcohol mixture induces the formation of a precipitate phase, which solidifies into an adsorbent membrane. After recovery, the membrane is rinsed to remove solvent traces and dried (Zheng *et al*., 2016). Coagulation-diffusion membrane formation utilises a coagulating polymer, such as polyethylene glycol (PEG) or PVA in a coagulation bath. The chitosan solution is immersed in this bath, where coagulation occur upon contact. The resulting membrane is subsequently rinsed and dried to eliminate residual solvents (González-Pabón *et al*., 2019). Additionally, an electroplating process can be employed, where an electrolyte containing dissolved chitosan is prepared and working electrodes, including platinum plates (Qu *et al.*, 2018) and titanium (Pantović Pavlović *et al.*, 2021) are used. Applying an electric current gradually deposits chitosan onto the electrodes, forming a chitosan membrane. This method allows for control over membrane thickness by adjusting electrochemical parameters such as deposition time and current intensity. Chitosan membranes share common limitations with other chitosan adsorbents, including low resistance to physical deformation, sensitivity to environmental conditions, and limited selective permeability, which affects their effectiveness depending on the compound (Wang *et al*., 2023).

Chitosan membranes and their derivatives have proven effective in removing dyes and heavy metal ions (Daraei *et al.*, 2013). For instance, Li *et al.* (2016) found that Al-doped chitosan non-woven fabrics were more effective in dye removal than plain chitosan non-woven ones due to chelation between the dyes and metal centres in the doped adsorbent fabric, achieving a maximum adsorption capacity of 260.03 mg/g. Their study highlighted the material's technical feasibility, reusability, and efficiency. Similarly, Karim *et al.* (2014) developed fully bio-based composite membranes for water purification using cellulose nanocrystals (CNCs) in a chitosan matrix, which effectively removed 98%, 84%, and 70% of Victoria blue 2B, methyl violet 2B, and rhodamine 6G, respectively, after 24 hours. Further innovations included Rashid *et al*. (2015), where chitosan-Cu-Fe bimetal complex/H_2O_2 system, which efficiently removed over 90% of dyes within 10 minutes

across a pH range of 4-12 and Makaremi *et al*. (2016), in which polyacrylonitrile nanofiber functionalised with zinc oxide nanoparticles (ZnO) enhanced the membrane's adsorption efficiency and mechanical strength. Mahmoud *et al*. (2017) designed a novel nanocomposite by cross-linking chitosan nanolayers onto TiO_2 nanoparticles, achieving an adsorption capacity of up to 1,150 μmol/g at various pH levels. Ma *et al.* (2017) synthesised a double network hydrogel using EDTA cross-linked chitosan and N, N-methylene bisacrylamide, demonstrating high adsorption capacities for Cd(II), Cu(II), and Pb(II). Advanced materials like cerium-functionalised PVA-chitosan nanofiber adsorbents have been developed for the removal of toxic ions such as Hg(II) from aqueous systems, achieving significant adsorption capacities. Ekambaram *et al*. (2017) modified PVDF nanofiltration membranes with carboxymethyl chitosan-zinc oxide nanocomposites, resulting in improved rejection of inorganic salts and reduced fouling rates. Sabourian *et al.* (2016) created a composite nanofibrous adsorbent using chitosan, tetraethyl orthosilicate and aminopropyl triethoxysilane, achieving high adsorption efficiencies for Ni(II), Cu(II), and Pb(II).

Fibres and Nanofibres

Chitosan in fibre form contributes significantly through its mechanical and structural properties. Like chitosan beads, chitosan fibres exhibit remarkable mechanical strength and flexibility, making them suitable for various applications that require robust and adaptable materials. The structural properties of chitosan fibres can be modified using diverse synthetic techniques. For example, electrospinning involves placing a chitosan solution in a syringe or reservoir and applying a high electrical voltage. The electric force drives the solution through a capillary (needle or nozzle), forming a polymer jet. Rapid solvent evaporation during ejection creates nanoscale chitosan fibres deposited on a substrate, resulting in a large specific surface area and high porosity (Bigogno *et al.*, 2022). This versatile technique allows precise control over fibre size, morphology, and orientation by adjusting process parameters. Alternatively, wet manufacturing methods use coagulation, where a chitosan solution is injected into a coagulation bath, leading to rapid solvent diffusion that solidifies the solution into cohesive chitosan fibres (Miranda *et al.*, 2023). The morphology of these fibres can be controlled through adjustments in the bath composition and subsequent treatments such as cross-linking (Rostamitabar *et al.*, 2022) or heat treatment (Mak & Leung, 2019). Jafri *et al.* (2023) explored enhancing fibre properties by combining chitosan with carboxymethylcellulose (CMC) to compensate for CMC's inherent low physical strength due to its high hydrophilicity. Chitosan fibres, as previously mentioned are particularly relevant in biomaterial applications within the medical sector. Perrin *et al.* (2022) demonstrated the use of fungal and shrimp-derived chitosan in suturing applications, highlighting their biocompatibility and mechanical resilience.

Foams and Sponges

Chitosan foams, commonly referred to as sponges, are three-dimensional materials distinguished by their spongy texture and porous architecture. These foams are produced using methods akin to those for adsorbent hydrogels, including freeze-drying (Deng *et al.,* 2023), gel foaming (Vo *et al.,* 2023), emulsification (Wang *et al.*, 2022), and supercritical CO_2 extraction (Song *et al.*, 2014). These techniques introduce air bubbles or pores into the chitosan matrix, forming the foam structure. Post-formation, the foams are subjected to drying processes such as freeze-drying again (Shimojo *et al.*, 2015), air drying (Chen *et al.*, 2015) or heat drying (Andrieux *et al.*, 2017) to remove solvents and stabilise the structure.

The porous architecture of chitosan foams imparts unique properties such as a high specific surface area and substantial adsorption capacity (Zhao *et al*., 2020). This structure facilitates efficient molecular diffusion, making these foams ideal for applications like adsorption, controlled

substance release (Platon *et al*., 2023) and tissue regeneration (Chen *et al.*, 2022). Chitosan foams are adaptable and can be customised for a range of uses in biotechnology, regenerative medicine, environmental applications, and functional materials (Xu *et al*., 2021). However, challenges such as low mechanical stability and moisture resistance persist. The mechanical fragility of chitosan foams is a notable limitation, attributed to their porous nature, which reduces density and compactness, rendering them susceptible to deformation and abrasion (Liu *et al.*, 2021). The pores can serve as stress points, leading to structural weaknesses, particularly when the pores are large or poorly interconnected (Chashchin *et al*., 2021). Strategies to enhance their mechanical strength include the incorporation of polymers, structural modifications, and reinforcement techniques (Ergun, 2023). Additionally, the porous structure and presence of amine groups make chitosan foams prone to moisture absorption, which can accelerate degradation in humid conditions (Lujan *et al*., 2022). This moisture absorption can undermine their dimensional stability and durability, especially in humid environments or when in contact with biological fluids (Ottenhall *et al*., 2018).

Chitosan's Potential in Removing Antibiotics

From 2008 to 2018, aquaculture in 11 major-producing countries saw the use of 67 different antibiotic compounds, a notable increase from the 27 drugs documented between 1990 and 2007 (Lulijwa *et al*., 2020). Despite efforts to regulate, banned antibiotics like chloramphenicol and ciprofloxacin have been detected in aquaculture products, underscoring the ongoing challenge of enforcement and monitoring in the industry (Lulijwa *et al.*, 2020).

Tetracycline

Tetracycline (TC) is extensively employed in marine aquaculture, with research showing that 50% to 80% of the administered antibiotic is excreted via the urine and faeces of aquatic organisms, leading to contamination of water bodies (Sarmah *et al*., 2006; Suzuki *et al*., 2019). This environmental and ecological risk highlights the urgent need for effective TC removal in aquaculture systems.

Zhang *et al*. (2016) developed magnetic core-brush composite adsorbents by grafting co-polymers onto chitosan/Fe_3O_4 composites (CS-MCP). These composites were found to be cost-effective adsorbents for TC removal from both single and binary solutions containing diclofenac sodium. The study showed that modifications using 2-methyl acryloyloxyethyl trimethyl ammonium chloride (CD-MCP), acrylic acid (CA-MCP), and methyl methacrylate (CM-MCP) could enhance TC adsorption performance. Among these, CD-MCP had the highest efficiency due to its positively charged surface, which exhibited a strong affinity for anionic species through electrostatic attraction. The presence of diclofenac sodium enhanced CD-MCP's adsorption capacity, likely due to strong π-π interactions with aromatic rings after adsorption onto the poly (2-methyl acryloyloxyethyl trimethyl ammonium chloride) branches.

In another research, Liu *et al.* (2019) developed a chitosan-Fe/S modified biochar (BCFe/S) for TC removal from water. BCFe/S showed superior adsorption ability compared with pristine chitosan, Fe/S or biochar alone. The adsorption performance varied significantly with the mass ratio of biochar, chitosan, and $FeSO_4$, with maximum adsorption capacities of 75.36 mg/g at pH 9 for biochar and 180.39 mg/g at pH 5 for BCFe/S. Addition of Na^+, K^+, NH_4^+, and Ca^{2+} negatively influenced TC adsorption onto BCFe/S due to ion exchange, chelation, hydrated radius, and hydrogen bonding effects, although Ca^{2+} did not significantly affect TC adsorption by biochar. Main mechanisms for TC removal by BCFe/S included electrostatic attraction, π-π stacking, pore filling, silicate bonding, hydrogen bonding, chelation, and ion exchange. Higher temperatures facilitated a more homogeneous biochar surface, whereas it was more heterogeneous for BCFe/S surface,

indicating different adsorption site behaviours. TC adsorption by BCFe/S and biochar may be classified as chemisorption and physisorption, respectively.

Industrial wastewater often contains pharmaceutical compounds and metal ions. High levels of TC and copper ions, for example, in animal waste, pose substantial environmental risks to aquatic systems. Studies have employed chitosan derivatives as flocculants for TC and Cu(II) ions. Carboxyethyl chitosan was used to flocculate solutions containing such pollutants, where charge neutralisation was the primary mechanism for Cu(II) removal and TC was synergistically removed by embedding in Cu(II) hydroxides. Jia *et al.* (2016) reported that modifying chitosan flocculants with 2,4-bis(dimethylamino)-6-chloro-(1,3,5)-triazine (BDAT), which contained aromatic rings could enhance the removal efficiency of TC and Cu(II) through strong interactions between BDAT-chitosan, Cu(II) and TC, including charge attraction and coordination between Cu(II) and BDAT-chitosan, coordination between TC and Cu(II), and π-π stacking between the negatively charged aromatic rings in TC and positively charged triazine rings in BDAT-chitosan.

In another study, Oladoja *et al*. (2014) highlighted the high adsorption efficiency of magnetic macro-reticulated cross-linked chitosan (MRC) synthesised using a gastropod shell as a pore-forming agent for TC removal. They found an adsorption energy of 100 kJ/mol, suggesting a chemisorption-dominated and kinetically controlled TC sorption process rather than being diffusion-controlled. The study also indicated that organics like humic acid could negatively impact TC removal performance. Huang *et al.* (2017) studied a magnetic Fe_3O_4@ SiO_2-chitosan/graphene oxide (MSCG) nanocomposite for TC adsorption, finding it to be an effective adsorbent through π-π and electrostatic interactions. They discovered that the presence of Cu(II) significantly enhanced TC adsorption by acting as a bridge between the analyte and adsorbent, increasing the maximum adsorption capacity from 67.57 mmol/kg to 183.47 mmol/kg. The adsorption was attributed to electrostatic interactions between positively charged nitrogen molecules in MSCG and TC, and strong π-π interactions between the benzene rings in TC molecules and the bulk π systems on the graphene oxide surface. The TC adsorption was further enhanced by oxygen-containing functional groups (C=O, –OH) on TC that showed strong affinity towards Cu(II). Addition of cations like Na^+, K^+, Ca^{2+}, and Mg^{2+} did not significantly affect TC adsorption. Kang *et al.* (2010) observed a similar effect with Cu(II) enhancing TC sorption onto chitosan, increasing the maximum adsorption capacity from 53.82 to 93.04 mmol/kg as copper concentration rose from 0 to 0.5 mmol/L, although TC presence negatively affected copper adsorption.

In 2009, Caroni *et al*. (2009) examined the adsorption of TC onto chitosan particles, identifying two main steps: Deprotonation of tetracycline hydrochloride, followed by protonation and adsorption of TC onto chitosan. Using the solution depletion method, they explored the adsorption of protonated TC on chitosan. Their results indicated that TC adsorption involved surface protonation of chitosan, which was directly related to the concentration in the continuous phase. Higher TC concentrations caused disruption of the chitosan surface, particularly at around 1.2 g/L, leading to increased TC adsorption. The adsorption kinetics before surface disruption showed different behaviours for protonated and non-protonated TC, with the latter playing a dominant role. The adsorption rate for protonated TC matched the surface charging rate while non-protonated TC exhibited a higher rate due to repulsive forces between protonated TC and the positively charged chitosan sites.

Abdolmaleki *et al.* (2018) investigated the use of glutaraldehyde-crosslinked electrospun nanofibers of chitosan/PVA as a novel adsorbent for TC. These nanofibers produced using a 75:25 volumetric ratio of chitosan to PVA had diameters ranging from 3-11 nm before and 6-18 nm after glutaraldehyde crosslinking. Response

surface methodology revealed that various parameters, including the volumetric ratio of PVA to chitosan, initial TC concentration, pH and adsorbent dosage, significantly influenced adsorption capacity, achieving a maximum of 102 mg/g.

Ma *et al.* (2019) demonstrated that carboxymethyl-chitosan amplified the interlayer spacing of Na-montmorillonite, improving its adsorption capacity for TC and chlortetracycline (CTC). Adsorption equilibrium was reached within two hours, facilitated by cation exchange and electrostatic interactions between negatively charged antibiotic anions and positively charged carboxymethyl-chitosan at pH 4 to 7. Li *et al.* (2020) studied a magnetic $NiFe_2O_4$-COF-chitosan-terephthalaldehyde nanocomposite film (NCCT) for TC and cefotaxime (CTX) adsorption. The synergetic effect of hydroxyl and amine groups of chitosan with the phenyl group of COF and terephthalaldehyde enhanced adsorption behaviour, with maximum adsorption capacities of 388.52 mg/g at pH 8 for TC and 309.26 mg/g at pH 4 for CTX. Adsorption mechanisms for TC included complexation, cation exchange, electrostatic attraction, hydrogen bonding and π-π interactions while CTX adsorption involved condensation reactions and similar interactions.

Metal-organic frameworks (MOFs) have gained attention as innovative adsorbents to capture TC molecules. Zhao *et al.* (2020) incorporated MOFs (ZIF-8, ZIF-67, HKUST-1, and Fe-BTC) into a chitosan matrix for TC adsorption, finding that ZIF-8-chitosan composite beads exhibited high adsorption capacity and reusability with a maximum of 495.04 mg/g. The ZIF-8-chitosan column could treat approximately 887 bed volumes of TC solution in fixed-bed experiments, utilising hydrogen bonding, electrostatic interaction, and π-π stacking interactions.

Recently, Ahamad *et al.* (2020) prepared a mesoporous magnetic chitosan/thiobarbituric acid/malondialdehyde-Fe_3O_4 nanocomposite (CTM@Fe_3O_4) with a high surface area of 376 m^2/g, which showed efficient TC adsorption with a capacity of 215.31 mg/g. The adsorption followed the Langmuir isotherm and pseudo-second-order kinetic model, driven by π-π conjugative effects and hydrophobic interactions due to phenyl rings and carbonyl groups in TC. Ranjbari *et al.* (2020) demonstrated that tricaprylmethylammonium chloride (TCMA) ionic liquid impregnation significantly improved chitosan's adsorption efficiency for TC. The chitosan-based materials were further modified with TCMA, enhancing the adsorption capacity and providing an effective method for removing TC from aqueous solutions.

Amoxicillin

Amoxicillin (AMX), a commonly used β-lactam antibiotic, disrupts cell wall peptidoglycan synthesis in actively growing bacteria by targeting enzymes involved in cross-linking such as transpeptidases and carboxypeptidases. β-lactam antibiotics' effectiveness relies on their enzyme-binding affinity, presence of bacterial capsule and peptidoglycan barriers, and β-lactamase stability, which is regulated via constitutive or inducible mechanisms (Aoki, 1997; Bruun *et al.*, 2000). AMX is widely used to treat Aeromonas-induced furunculosis in salmonids, administered orally through medicated feed at 80-160 mg/kg/day for 10 days (Ang *et al.,* 1996). Environmental persistence of β-lactam antibiotics is generally low, as they are prone to biological and physicochemical degradation in the environment due to their nature as amino acid-based synthetic metabolites.

However, AMX may persist in the environment and pose significant risks to various organisms due to its limited degradability (Putra *et al.*, 2009). Adriano *et al.* (2005) conducted a seminal study using chitosan beads to adsorb AMX in batch experiments, achieving an adsorption capacity of 8.71 mg/g. But the rigid chitosan matrix significantly impeded AMX diffusion into beads, limited by the molecular diffusivity of AMX in water. Another investigation employed chitosan-stabilised bimetallic Fe/Ni nanoparticles (CSeFe/Ni) to simultaneously remove AMX and Cd(II) ions

from aqueous solutions (Weng et al., 2013). This approach initially adsorbed AMX and Cd(II) onto the CSeFe/Ni surface, followed by catalytic AMX degradation (activation energy of 60.9 kJ/mol) through β-lactam bond cleavage. Despite effective catalysis, iron oxide formation and corrosion on the CSeFe/Ni surface eventually led to Fe/Ni leaching, reducing adsorbent reactivity and AMX removal efficiency from 68.9% to 2.2% over three cycles (Weng *et al.*, 2013). The CSeFe/Ni nanoparticles demonstrated removal efficiencies of 63% for AMX and 79% for Cd(II) in aquaculture wastewater.

Erythromycin

Erythromycin, produced by *Streptomyces erythreus*, acts by binding to the bacterial ribosome 50S subunit, disrupting protein synthesis and demonstrating heightened effectiveness against Gram-positive bacteria due to steric interactions. It is particularly effective in treating bacterial kidney disease (BKD) caused by *Renibacterium salmoninarum* in salmonids. Typically administered through feed at a dosage of 50-100 mg/kg body weight per day for approximately 21 days, this regimen has been shown to reduce BKD mortality in brook trout by 50% (Stoffregen *et al*., 1996). Despite lacking approval from the International Council for the Exploration of the Seas (ICES), erythromycin was found to exhibit excretion levels of 0.03-0.08 mg/g in yellowtails 168 hours after a treatment regimen of 50 mg/kg/day for five days (Stoffregen *et al.*, 1996). Acute toxicity was minimal at doses exceeding 2 g/kg, with no observed abnormalities, although rainbow trout exposed to 100 mg/kg/day for 21 days displayed behavioural and physiological abnormalities. Concerns regarding erythromycin's environmental impact stem more from antibiotic resistance than persistence, as its ether linkages are susceptible to reduction or oxidation through physicochemical or biological processes.

Ou *et al.* (2015) innovatively developed a chitosan-stabilised Pickering emulsion enhanced with magnetic imprinted polymers for erythromycin removal. Their study demonstrated that erythromycin exhibited a higher affinity for adsorption onto the magnetic polymer compared with chloramphenicol and azithromycin, achieving an impressive adsorption capacity of 52.32 mmol/g at 15°C while maintaining high regeneration potential (Ou *et al*., 2015).

Norfloxacin

Norfloxacin (NOR), a widely used antibacterial drug in Asian countries (Wiwattanapatapee *et al.*, 2002) is among the antibiotics with the highest residual levels in China's aquaculture industry. Its significant presence, accounting for 12% of total cases is likely due to its relatively higher rate of use (Liu *et al*., 2017). NOR, a notable antibiotic has been detected in the effluents of wastewater treatment plants and poses toxicity risks to organisms (Meler *et al.*, 2012; Wammer *et al.*, 2013). Chitosan has shown effectiveness in adsorbing chemotherapeutic NOR under simulated in vitro conditions, achieving removal efficiencies of between 80% and 98% (Meler *et al.*, 2012). Wu *et al.* (2016) employed molecularly imprinted γ-Fe_2O_3 cross-linked chitosan (MICs) for targeted NOR removal, utilising hydrogen bonding and van der Waals forces between NOR and the adsorbents. Their study highlighted MICs' ability to selectively adsorb NOR in the presence of competitive organic matter like phenol, ofloxacin, and sulphadiazine. This selectivity was attributed to specific binding sites for pharmaceutical molecules and intermolecular hydrogen bonding. The molecular imprinting mechanism effectively reduced humic and fulvic acid-like organics, while enhancing the adsorption of tryptophan-like substances. They found Fenton-like oxidation to be superior for regeneration compared to NaOH washing, preserving more binding sites and facilitating oxidative breakdown of desorbed NOR.

In a recent study, Zhou *et al.* (2018) developed core-brush shaped chitosan/Fe_3O_4 composites (CS-MCPs) with numerous functional groups on the branches. Innovative modifications to chitosan-based composites with polyanion branches led to remarkable adsorption capacities, reaching 165 mg/g for

NOR and 134 mg/g for tylosin [poly(sodium p-styrenesulphonate)]. Conversely, polycation branches with modified CS-MCPs were preferred for diclofenac sodium adsorption, achieving a capacity of 151 mg/g [poly(p-vinylbenzyl trimethylammonium chloride)].

Chloramphenicol

Chloramphenicol (CAP) is a synthetic antibiotic banned by the European Union for use in food-producing species due to its severe toxicity to human health (Sun *et al.,* 2017). Its use in livestock destined for human consumption has been heavily restricted (Nicolich *et al*., 2006; Munns *et al.*, 2020), leading to its prohibition in many countries for treatment of food-producing animals, including those in aquaculture. Despite this ban, CAP continues to be utilised in some developing nations due to its low cost (Mottier *et al*., 2003). While aquatic animals in aquaculture can metabolise CAP extensively, the potential for residual contamination in their bodies and the environment remains a significant concern. Its highly polar nature leads to a low affinity for solid surfaces (Sun *et al*., 2017). To enhance CAP adsorption from aqueous solutions, Ma *et al.* (2015) developed magnetic chitosan microspheres to create molecularly imprinted (MIPs) and non-imprinted polymers (MNIPs), concluding that MIPs demonstrated significantly higher CAP adsorption activity compared with MNIPs, with CAP being selectively bound to the surface at a capacity of 27.5 μmol/g (Ma *et al.*, 2015). This selective binding was observed even in the presence of other common antibiotics with adsorption capacities of 6.6, 15.1, and 24.8 mg/g, respectively. Additionally, Meler *et al.* (2013) investigated the binding capacity of CAP on chitosan under pharmaceutical conditions in vitro using a dynamic model. Their research indicated that the binding capacity of chitosan towards CAP is influenced by factors such as CAP concentration, viscosity, pH of the environment, and the compounds present in the medium. They reported similar findings for the adsorption of clarithromycin (Meler *et al.*, 2013).

Ciprofloxacin

Ciprofloxacin (CIP), a critical antibiotic effective against both Gram-negative and Gram-positive bacteria in human and veterinary medicine is essential for treating severe bacterial infections. Despite its importance, CIP has faced challenges due to its overuse and misuse, leading to the development of antibiotic resistance. It was initially developed in the 1980s, with modifications that enhanced its broad-spectrum capabilities by substituting the ethyl group with a cyclopropyl group. CIP targets bacterial DNA gyrase and topoisomerase IV, disrupting vital processes like DNA replication and repair (Drlica, 1999).

Despite being banned in certain contexts due to concerns over antibiotic resistance and environmental impact, ciprofloxacin continues to be used, contributing to its widespread presence in municipal sewage and wastewater plant effluents. This persistence underscores its role in environmental contamination and the ongoing challenges in regulating its use to prevent the proliferation of antibiotic resistance. Recent findings have highlighted elevated levels of CIP in landfill leachate in Hong Kong (Chung *et al.,* 2018), indicating its presence beyond regulated environments and raising concerns about its impact on public health.

Cross-linked chitosan-metal microspheres such as chitosan-Fe(III) and chitosan-Zn(II) have been shown to exhibit superior adsorption capabilities for CIP compared with chitosan and chitosan-Fe(II) microspheres (Reynaud *et al.,* 2011). Innovative approaches include the synthesis of magnetic chitosan grafted graphene oxide (MCGO) nanocomposites by Wang *et al.* (2016), which demonstrated enhanced adsorption of CIP (282.9 mg/g) due to π-π electron interactions and electrostatic attractions. However, the adsorption capacity of MCGO was found to decrease significantly with increased ionic concentrations (NaCl and CaCl2), suggesting a dependency on environmental conditions for optimal performance (Wang *et al.*, 2016). Recently, Rasoulzadeh *et al.* (2019) developed magnetite imprinted chitosan polymer

nanocomposites (Fe-CS NCs) for CIP removal, showing pH-dependent adsorption mechanisms. At pH levels around neutral, electrostatic attractions and hydrophobic interactions were most effective while at extremes (pH < 5.9 or > 8.9), electrostatic repulsion reduced efficiency (Rasoulzadeh *et al*., 2019). This innovative use of Fe-CS NCs highlights ongoing efforts to refine adsorption technologies for mitigating CIP contamination in aquatic environments.

Sulphanilamide

Sulphanilamide, a widely utilised broad-spectrum antibiotic for treating protozoan and bacterial infections, poses significant environmental risks due to its persistent residues (Tian *et al.*, 2020). Recently, researchers have developed innovative approaches to mitigate its impact, such as the use of a hierarchical porous starch-chitosan-UiO-66-COOH composite for treating sulphanilamide-contaminated pharmaceutical wastewater (Jia *et al.*, 2021). This composite leverages the stable macro-porous structure of starch to host UiO-66-COOH nanoparticles, with chitosan enhancing adsorption through hydrogen bonding and electrostatic interactions. The Zr-O groups in the carboxylic Zr-MOF composite play a pivotal role in chemisorbing sulphanilamide, demonstrating high affinity via a chemisorptive process.

The adsorption capacity of the starch-chitosan-UiO-66-COOH composite for sulphanilamide declines as solution pH increases from 3 to 11, which may be attributed to increased anionic species in acidic conditions. Kinetic studies indicate that sulphanilamide adsorption follows a pseudo-second-order process while adsorption isotherm modelling fits well with the Langmuir model, confirming a homogeneous monolayer chemisorption mechanism. These advancements underscore ongoing efforts to develop effective strategies for mitigating sulphanilamide contamination in wastewater treatment processes.

Conclusions

Chitosan-based materials demonstrate considerable promise for the removal of antibiotics and emerging contaminants from aquaculture wastewater. The studies reviewed showcase diverse approaches and innovative modifications of chitosan-based materials. Through these approaches, the chitosan had enhanced adsorption efficiencies by leveraging chemical interactions and surface modifications. These advancements highlighted the versatility and efficacy of chitosan in addressing the growing concerns of antibiotic contamination, offering sustainable solutions to mitigate environmental risks and safeguard aquatic ecosystems. The selective adsorption capabilities of molecularly imprinted chitosan and pH-dependent mechanisms further underscored its versatility in tackling diverse antibiotic pollutants. Moving forward, continued research and development in this area would be crucial to optimise adsorption capacities, improving regeneration methods and understanding the underlying mechanisms that were crucial for the sustainable management of antibiotic residues in aquatic environments.

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Conflict of Interest Statement

The authors declare that they have no conflict of interest.

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