RSC Advances

REVIEW

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View Journal | View Issue

Cite this: *RSC Adv.*, 2025, **15**, 20168

Removal of pharmaceutical residues from aquatic systems using bimetallic metal–organic frameworks (BMOFs): a critical review

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Received 30th April 2025
Accepted 10th June 2025

DOI: 10.1039/d5ra03056k

rsc.li/rsc-advances

In recent years, pharmaceuticals have become a major environmental issue due to their ongoing release and persistence in aquatic ecosystems, even at low concentrations. Among various solutions, bimetallic metal–organic frameworks (BMOFs) have attracted considerable attention. This is not only because of their tunable pore structures, large surface area, and excellent reactivity but also due to the incorporation of multiple metal ions, which enhance their ability to remove and degrade pharmaceutical residues. This review provides a detailed analysis of the advantages of BMOFs, introduces the occurrence of pharmaceutical residues and their toxic effects on the environment and humans, and, for the first time, explores their applications in removing pharmaceutical residues. Additionally, we discuss current challenges and future perspectives for BMOFs, aiming to advance their development and maximize their potential in environmental applications. We aim to provide detailed and meaningful insights to researchers in both materials science and environmental studies, thereby driving advancement in this interdisciplinary arena.

1 Introduction

Pharmaceutical pollutants are increasingly building up in both wastewater and freshwater systems, emerging as a significant environmental concern. Each year, around 300 million tons of pharmaceutical and industrial chemicals are discharged into natural water sources.[1] These micropollutants pose serious environmental hazards due to their non-biodegradable nature, high toxicity, and unique molecular structures.[2] Consequently, it is crucial to remove pharmaceuticals from wastewater to mitigate their harmful impacts on the environment, human health, and aquatic ecosystems.[3–5] Aquatic environments are continuously exposed to these persistent contaminants because wastewater treatment processes often fail to remove them completely. Pharmaceuticals are typically excreted *via* urine and feces as parent compounds, metabolites, or conjugates with glucuronic and sulfuric acids.[6] These substances enter aquatic ecosystems through the discharge of both treated and untreated wastewater.

The presence of pharmaceuticals in water can be linked to sources such as personal care products, waste from the pharmaceutical industry, hospital waste, and therapeutic drugs. The detection of trace amounts of pharmaceuticals and other xenobiotic compounds in treated drinking water raises significant public health concerns. Limited knowledge exists about the potential chronic health effects of long-term exposure to these compound mixtures through drinking water.[7–9] Therefore, addressing the removal of pharmaceuticals and other priority pollutants from wastewater before discharge has become a critical issue in environmental science. Significant efforts are needed to study this problem and mitigate its impacts effectively.

Recent studies emphasize photocatalytic degradation of pharmaceuticals for its affordability and eco-friendliness.[10] Materials such as metal oxides,[11] activated carbon,[12] , and metal–organic framework (MOF)-based nanoparticles have gained significant attention for their success in environmental treatment and protection.[13] MOFs, in particular, have drawn interest because of their tunable cavities, crystalline structures, high surface area, open framework, and diverse designs achieved through the combination of sources of metal ions and organic linkers.[14–20] Their high specific surface area and easily tunable porous structures make MOFs increasingly attractive for catalysis. In various applications, MOFs serve as catalysts and adsorbents for removing pollutants from wastewater.[21]

Compared to monometallic MOFs, bimetallic MOFs (BMOFs) offer enhanced and distinct functionalities. The incorporation of two metal ions increases the number of active sites, improving structural stability and catalytic efficiency due to synergistic interactions.[22] BMOFs are beneficial as multivalent metals provide extra redox-active sites.[23,24] Some BMOFs are created by altering the synthesis of existing MOFs to include a second metal ion. Ligand–metal interactions with similar electronic properties promote a single-phase bimetallic structure over separate monometallic compounds.[25] These

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heterometallic derivatives retain similarities to monometallic MOFs but often exhibit greater efficiency, unique properties, and stability.[26,27] Therefore, it is crucial to develop BMOFs with enhanced activity and to extensively explore their potential applications in removing residual pharmaceuticals.

Numerous research groups have explored different approaches to treating wastewater contaminated with pharmaceutical pollutants using various materials. This review summarizes the application of BMOFs in removing pharmaceutical pollutants, highlighting their efficiency as catalysts and adsorbents compared to other materials. Additionally, it examines the presence of pharmaceutical residues, their harmful environmental and human health effects, and the potential future applications of BMOFs in this field. To the best of our knowledge, no existing review comprehensively focuses on the removal of pharmaceutical pollutants specifically using BMOFs. Therefore, this paper aims to provide the most up-to-date insights into the use of BMOFs for treating pharmaceutical-contaminated wastewater, along with future research directions.

2 Occurrences of pharmaceutical residues and their toxic effects on the environment and humans

The widespread presence of xenobiotics in the environment has raised increasing concerns among urban authorities and health professionals due to their persistence and extended half-life. In recent decades, rapid industrial growth and urban expansion have led to the excessive depletion of essential natural resources.[28] The manufacturing processes for consumer goods involve several stages that release significant amounts of waste in various forms liquid, solid, or gaseous forms, all of which pose environmental hazards.[29,30] According to the World Health Organization (WHO), around 15% of hospital waste is categorized as infectious, making it both toxic and dangerous. Wastewater has long been a major reservoir for pharmaceutical compounds (PCs),[31] personal care products,[32] pesticides,[33] and other similar pollutants. The widespread presence of PCs in the environment can be attributed to their persistent release and slow transformation rates. Pharmaceutical manufacturing industries generate wastewater containing a diverse array of PCs, often with high chemical oxygen demand (COD) and occasionally elevated salinity levels.[34] The minimum concentrations of PCs in aquatic environments have been reported to range from ng L^{-1} to μg L^{-1}.[35] PCs are chemically stable and frequently referred to as micropollutants due to their complex fate and transport mechanisms. The physicochemical properties of these compounds play a critical role in determining the extent and severity of their contamination across various environmental media, including soil, water, and air. Between 2000 and 2015, global antibacterial medication usage increased by approximately 65%, and pharmaceutical consumption worldwide is projected to rise by 200% by 2030, compared to the 42 billion defined daily doses recorded in 2015.[36] The COVID-19 pandemic further accelerated the unprecedented use of drugs and medications.[37] Veterinary pharmaceuticals are often excreted directly onto the ground or into surface waters without undergoing treatment at wastewater treatment plants (WWTPs), making their management and monitoring significantly more difficult. The soil can serve as a major source of water pollution,[38] as many of these substances and their metabolites are water-soluble and are expelled through urine and feces.[39] In intensive livestock farming, these pharmaceuticals may enter the environment indirectly through the use of manure and slurry as fertilizers, potentially transferring to humans *via* the food chain. Additionally, pharmaceuticals utilized in fish farming are released directly into surface waters.[39]

The pharmaceuticals most commonly detected in water treatment effluents include steroids, antidepressants, antibiotics, antacids, analgesics, lipid-lowering agents, tranquilizers, anti-inflammatory drugs, antipyretics, beta-blockers, and stimulants. PCs tend to infiltrate host environments, such as surface water, groundwater, the cryosphere, and wastewater, eventually transforming into intermediate products through interactions with biotic and abiotic environmental components. These transformations, influenced by the reactivity and sensitivity of PCs, pose significant threats to aquatic ecosystems, including the development of resistant microbial species.[40]

Pharmaceuticals in the environment pose significant risks due to their ecological toxicity, physicochemical properties, and consumption rates. Risk assessments are essential as these substances can bioaccumulate, exhibit high water solubility, persist in ecosystems, and potentially cause harmful or carcinogenic effects on organisms.[41] Even trace levels of pharmaceutical residues in the environment can lead to acute and long-term impacts on microbes, plants, and animals. These effects may range from metabolic disruptions to hormonal imbalances and can harm non-target species.[42] The complexity of pharmaceutical mixtures in the environment means that certain compounds can cause severe damage even at very low concentrations, sometimes below detectable thresholds. Some pharmaceuticals exhibit effects on non-human species similar to their effects on humans,[43] as they are designed to interact with specific receptors in humans and animals, which may also exist in other organisms. This interaction can inhibit essential biological processes such as cell envelope synthesis, protein synthesis, and nucleic acid synthesis.[44] Pharmaceuticals in the environment severely impact a wide range of organisms, with environmentally beneficial microorganisms being more affected than aquatic organisms. Drugs like fluoxetine, diclofenac, ibuprofen, and carbamazepine exhibit carcinogenic effects even at low concentrations, and diclofenac specifically causes acute kidney failure in humans and other toxic effects. The "complex pools" of pharmaceutical mixtures in nature often have greater toxicity than individual compounds, yet chronic effects at ecological levels remain underreported.[45] Long-term, low-dose exposure to pharmaceuticals in drinking water and their entry into the food chain through plants, vegetables, and meat raise concerns about cumulative impacts on health. Wastewater effluents contain measurable concentrations of harmful pharmaceuticals, which pose risks to microbes, humans, and higher organisms. Studies show that while some aquatic species tolerate acute toxicity from drugs, phytoplankton and invertebrates are particularly vulnerable. Chronic toxicity from compounds such as fluoxetine, carbamazepine, and 17 beta-estradiol presents significant risks to aquatic ecosystems, underscoring the need for further research on pharmaceutical mixtures and their ecological consequences.[46]

To reduce the presence of human and veterinary pharmaceuticals in the environment, it is essential to identify, test, and implement measures across short-, medium-, and long-term timeframes. Short-term measures to reduce pharmaceutical pollution focus on controlling emissions from production facilities, particularly in developing countries, and improving hygiene standards in hospitals and livestock farming to minimize infections and unnecessary antibiotic use. Efforts include promoting targeted antibiotic use, optimizing farm management practices, and exploring techniques such as manure treatment and biogas fermentation to reduce veterinary pharmaceutical residues. Mid-term measures emphasize developing environmentally friendly drugs, sustainable manufacturing processes, and formulations that minimize environmental impact. Long-term strategies involve designing eco-friendly pharmaceuticals through drug redesign and personalized medicine, supported by incentives for manufacturers to prioritize environmental sustainability. Additionally, enhanced monitoring, research on distribution pathways, and public education on proper drug disposal are essential across all timelines.[47] Fig. 1 provides a comprehensive overview of the various sources of pharmaceutical residues entering water systems, including industrial discharge, hospital waste, and agricultural runoff. It also illustrates their impact on wastewater contamination, soil pollution, potential health risks to humans, and adverse effects on aquatic ecosystems.

3 Bimetallic MOFs advancement over monometallic MOFs

MOFs are an emerging class of highly structured crystalline materials that form through the self-assembly of metal clusters and organic linkers *via* precisely coordinated bonds. Their unique physical and chemical properties have led to extensive applications in pollutant removal.[48,49] However, conventional MOFs encounter several challenges, including complex preparation processes, limited adsorption sites, structural instability, and the reliance on expensive metal salts. Research indicates that a high density of active metal sites significantly enhances pollutant adsorption capacity.[50] Compared to monometallic compounds (MMCs), BMOFs provide several benefits, including enhanced electrical conductivity, a greater number of active sites, adjustable electrochemical properties, and higher charge storage capacity. Integrating MOFs with other electrochemically active materials results in advanced composites with large specific surface areas, improved conductivity, and superior dispersion characteristics. Notably, certain BMOFs demonstrate increased electrocatalytic performance when exposed to light, making them suitable for use as photoelectrocatalysts.[51]

As a targeted strategy, BMOFs have garnered considerable attention across various fields.[52–55] These materials can be produced by altering the synthesis process of a particular MOF to incorporate a second metal ion. In this approach, the interaction between ligands and two metal ions with comparable electronic structures and charge distributions promotes the formation of a single-phase BMOF rather than a simple mixture of two separate MMCs.[23] Although these heterometallic derivatives share similarities with monometallic MOFs, they often demonstrate enhanced stability, efficiency, and other distinctive properties. This class of materials can function directly as electrode components or serve as templates or precursors in the fabrication of advanced composites.[56] BMOFs offer greater stability and efficiency than monometallic MOFs, enabling a dual-function mechanism or synergistic metal interactions.[26,57] The combination of two metal cations improves conductivity and enhances oxidation reactions, boosting electrocatalytic efficiency. It is evident that the incorporation of dual metal sites within a given MOF can result in superior electrochemical activity, attributable to the differing oxidation

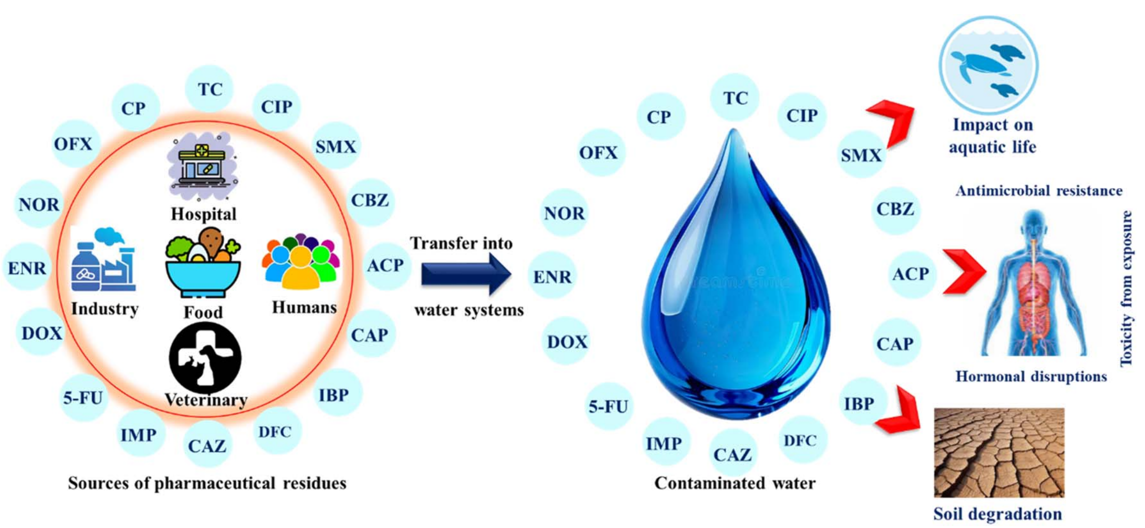

Fig. 1 Highlights the sources of pharmaceutical residues in water and their impact on wastewater, soil, humans, and aquatic life.

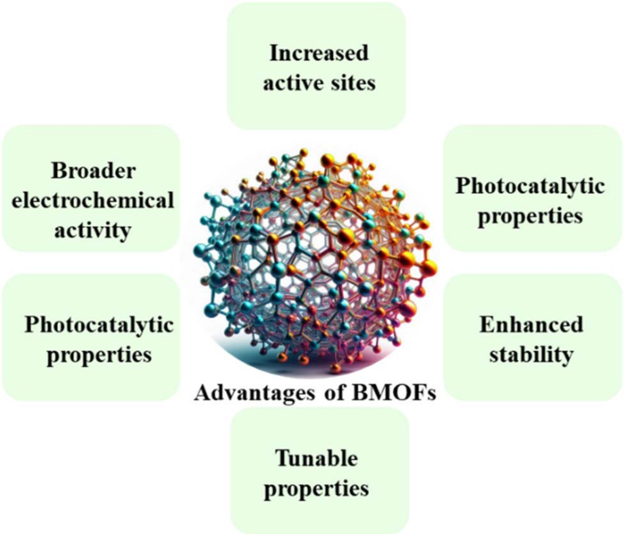

Fig. 2 The main unique characteristics of BMOFs.

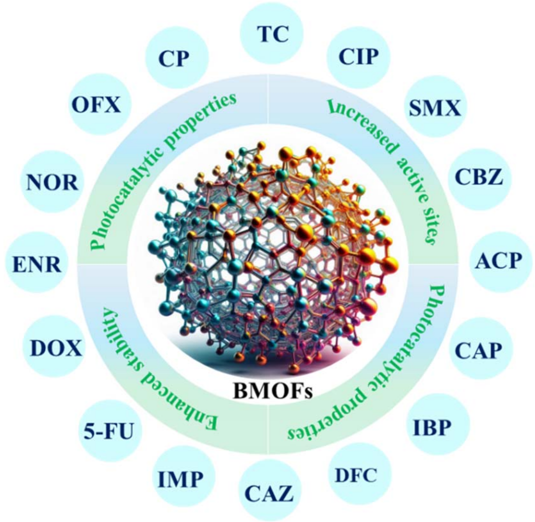

Fig. 3 Pharmaceutical residues removed using BMOFs.

potentials and associated electronic configurations.[58] Finally, facilitating multi-functional applications, BMOFs can serve multiple roles, such as functioning both as catalysts and adsorbents, making them suitable for integrated applications in environmental remediation.[59] Fig. 2 shows the main unique characteristics of BMOFs.

4 Applications

Bimetallic organic frameworks (BMOFs) and their derivatives demonstrate significant potential as alternative sorbents and catalysts for the efficient removal of a wide range of pharmaceuticals from aquatic solutions, including antibiotics, nonsteroidal anti-inflammatory drugs, and veterinary medications. The pharmaceuticals selected and reviewed in this study primarily include various classes, such as tetracycline (TC), ciprofloxacin (CIP), sulfamethoxazole (SMX), carbamazepine (CBZ), acetaminophen (ACP), chloramphenicol (CAP), ibuprofen (IBP), diclofenac sodium (DCF), ceftazidime (CAZ), imatinib (IMB), doxorubicin (DOX), 5-fluorouracil (5-FU), enrofloxacin (ENR), norfloxacin (NOR), moxifloxacin (MOX), cefoperazone (CP), sertraline, ofloxacin (OFX), and cefradine. As illustrated in Fig. 3.

4.1 Tetracycline removal

Since the discovery of antibiotics, bacterial infection-related diseases have been effectively managed. Among these, TC antibiotics, known as multifunctional broad-spectrum antibiotics, are widely utilized to treat bacterial infections in both humans and animals due to their ability to inhibit bacterial protein synthesis.[60] Additionally, TCs are used as feed additives in animal husbandry to promote animal growth. However, the overuse of antibiotics has led to a range of severe consequences.[61,62] Although TCs are biodegradable, their residual presence can cause selective genetic variations in microorganisms, resulting in the emergence of drug-resistant pathogens. Bacteria can acquire antibiotic resistance genes through mutations or gene transfer, and the exchange of these genes between agricultural soil bacteria and clinical pathogens often facilitates the spread of antibiotic resistance, giving rise to "superbugs". Recent studies have revealed that over 30 antibiotics, including various TCs and quinolones, have been detected in karst river systems, posing threats to non-target organisms across different trophic levels, such as algae, plants, bacteria, invertebrates, and fish.[63,64] However, the excessive use of TC can have significant impacts on human health, as TC residues may accumulate in foods such as meat and milk. Studies have shown that frequent consumption of TC can lead to liver damage and kidney issues in humans. Pregnant women are particularly vulnerable to TC-induced liver toxicity. Additionally, extensive data indicates that prolonged and repeated use of TC can negatively affect dental health by disrupting tooth growth and formation, as well as causing discoloration, turning teeth yellow.[65] BMOFs are promising porous materials for addressing environmental pollution caused by pharmaceuticals like TC, owing to their exceptional surface area, catalytic activity, and porous architecture. The following section reviews recent studies focused on the removal of TC, one of the most commonly used antibiotics, using BMOFs.

Chen *et al.* successfully synthesized bimetallic MOFs (MIL-53(Fe, Al)) for the efficient removal of TC from aqueous solutions. Their experiments on adsorption and photocatalysis revealed that a 3 : 2 molar ratio (40% MIL-53(Fe, Al)) yielded optimal performance. The adsorption process followed the Freundlich isotherm model and pseudo-second-order kinetics, with a maximum adsorption capacity of 402.033 mg g^{-1}. Under photocatalytic conditions, 10 mg of 40% MIL-53(Fe, Al) removed 94.33% of TC from a 70 mL solution (20 mg L^{-1}) within 50 minutes of irradiation, outperforming MIL-53(Fe) (71.39%) and MIL-53(Al) (81.82%). Additionally, the material demonstrated a strong adsorption-photocatalytic synergy, with the pseudo-first-order kinetic constant increasing by 3.11 times under direct irradiation without prior dark adsorption.[66] In another study, Xia *et al.* developed a BMOF gel (JLUE-MOG-Fe/Y) for the adsorption of chlortetracycline hydrochloride (CTC) from water. This material exhibited exceptional stability, adaptability, and recyclability, achieving a remarkable adsorption capacity of 584.83 mg g^{-1} at 25 °C.[67] Similarly, Zhang *et al.* designed transition metal/nitrogen-codoped hierarchically porous carbons (MNHCs) by pyrolyzing bimetallic ZIFs to enhance the adsorption of TC. The optimized MNHC, synthesized at 1000 °C with a 2% Fe/(Fe + Zn) molar ratio, featured a large specific surface area (920.73 m^2 g^{-1}), a hierarchical pore structure, high nitrogen content, and abundant Lewis acid sites. These properties significantly improved TC adsorption affinity and reduced diffusion resistance, leading to superior performance.[68]

Zhang *et al.* developed a novel Fe/Mn-MOF combined with a SnS_2 Z-scheme heterojunction photocatalyst through self-assembly. Leveraging the synergistic effects of the interfacial heterojunction, the photocatalyst demonstrated exceptional catalytic performance. With the aid of a persulfate-based advanced oxidation process, it achieved a degradation efficiency of nearly 91.4% for TC.[69] Lastly, Liu *et al.* fabricated Fe-doped zeolitic imidazolate frameworks-8 loaded cellulose (Fe/ZIF-8@cellulose) aerogels. The incorporation of Fe into ZIF-8 resulted in a maximum TC adsorption capacity of 1359.2 mg g^{-1}, surpassing the performance of previously reported ZIF-8-based polysaccharide adsorbents.[70]

Table 1 provides a summary of studies that have employed BMOFs for TC removal.

4.2 Ciprofloxacin removal

CIP one of the most commonly used second-generation quinolones, is widely employed in the treatment of bacterial infections.[100] However, CIP is often released into the environment through wastewater discharges and is frequently detected in various aquatic ecosystems. Wastewater from pharmaceutical industries and hospitals is particularly concerning, as it can contain extremely high levels of CIP contamination, reaching up to 31 mg L^{-1}.[101,102] Consequently, developing environmentally and economically sustainable methods to remove CIP from water is essential to mitigate public health risks associated with the emergence of antibiotic resistance in the environment.

Li *et al.* successfully synthesized a novel hetero-photo-Fenton (PF) catalyst, consisting of dual MOF-derived Fe–Zr oxide embedded in porous carbon skeleton. This hybrid photocatalyst, featuring a high surface area, well-developed porous structures, strong light absorption, and a narrow band gap, exhibited exceptional photo-Fenton activity, achieving around 99.1% degradation of CIP. Additionally, the catalyst system performed well in treating real water matrices.[103]. Lastly, Zhang *et al.* synthesized Cu/Ni-MOF for the targeted degradation of CIP in advanced oxidation processes (AOPs). The specific

Table 1 Removal of TC using BMOFs

Types of BMOFs	Method	Catalyst dosage	Initial concentration	Performance%	Ref.
Bi/Ni-MOF	Degradation	30 mg	10 mg L^{-1}	93.6	71
Fe/Ni-MOF	Degradation	20 mg	20 mg L^{-1}	95.76	72
Co/Zn-ZIF	Adsorption, degradation	60 mg L^{-1}	20 mg L^{-1}	89.54	73
Sn/Bi-MOF	Degradation	40 mg	20 mg L^{-1}	96.2	74
Ag/Bi-MOF	Degradation	30 mg	20 mg L^{-1}	83	75
Fe/Mn-MOF	Degradation	0.2 g L^{-1}	20 mg L^{-1}	90.95	76
Fe/Co-MOF	Degradation	50 mg/L	50 mg L^{-1}	91.76	77
Co/MIL-68(In)-NH_2	Degradation	0.6 g L^{-1}	10 mg L^{-1}	90.1	78
Fe/Co-MOF	Degradation	0.2 g L^{-1}	50 mg L^{-1}	99	79
Nb/Co-MOF	Degradation	0.2g L^{-1}	40 mg L^{-1}	97.8	80
Fe/Co-MOF	Adsorption	20 mg	100 mg L^{-1}	98	81
Fe/Co-MOF	Degradation	0.125 g L^{-1}	50 mg L^{-1}	90	82
Fe/Co-MOF	Adsorption, degradation	30 mg, 10 mg	70 mg L^{-1}, 20 mg L^{-1}	87.5, 91	83
Ni/Fe-MOF	Adsorption	1 g L^{-1}	400 mg L^{-1}	—	84
Fe/Bi-MOF	Degradation	0.5 g L^{-1}	20 mg L^{-1}	99.9	85
Fe/Co-MOF	Degradation	0.1 g L^{-1}	10 mg L^{-1}	100	86
Zr/Cu-MOF	Degradation	40 mg	—	94	87
Ni/Ti-MOF	Degradation	0.2 g L^{-1}	50 mg L^{-1}	83	88
Zn/Fe-MOF	Adsorption	10 mg	300 mg L	—	89
Co/Cu-MOF	Degradation	0.1 g L^{-1}	20 mg L^{-1}	98.7	90
Fe/Zn-ZIFs	Degradation	0.4 g L^{-1}	50 mg L^{-1}	92	91
Cu/Co-MOFs	Adsorption	50 mg	30 ppm	93.7	92
Fe/Co-MOF	Adsorption	—	20 mg L	—	93
Cu/Fe-ZIF-8	Adsorption	100 mg L	100 mg L	87.2	94
Zn/Cu-MOF-74	Adsorption	15 mg	30 mg L^{-1}	—	95
Zr/Fe-MOF	Degradation	10 mg	50 mg L^{-1}	87	96
Zn/Cu-MOF	Adsorption	20 mg	20 mg L^{-1}	96.55	97
Fe/Cu-MOF	Degradation	0.6 g L^{-1}	20 mg L^{-1}	93	98
Fe/Co-MOF	Degradation	10 mg	20 mg L^{-1}	93.34	99

Table 2 Removal of CIP using BMOFs

Types of BMOFs	Method	Catalyst dosage	Initial concentration	Performance%	Ref.
Fe/Cu or Mn-MOF	Degradation	0.1 g L^{-1}	20 mg L^{-1}	88.96	105
Cu/Co-MOF	Degradation	25 mg	20 mg L^{-1}	90	106
Ti/Bi-MOFs	Degradation	20 mg	10 mg L^{-1}	93.3	107
Zn/Co-ZIF	Degradation	0.1 g L^{-1}	20 mg L^{-1}	90	108
Zn/Co-ZIF	Adsorption	0.5 g L^{-1}	—	85.30	109
In/Cu-MOF	Degradation	2 mg	15 mg L^{-1}	81.70	110
Ce/Zr-MOF	Degradation	20 mg	20 ppm	90.8	111
Fe/Cu-MOF	Adsorption, degradation	0.1 g L^{-1}	15 mg L^{-1}	74.48, 57.88	112
Fe/Mn-MOF	Degradation	5 mg	20 mg L^{-1}	98.3	113

recognition sites on Cu/Ni-MOF, enabled by electrostatic interactions and functional group binding with CIP, provided excellent selective recognition ($Q_{\max} = 14.82$ mg g^{-1}). This allowed active radicals to efficiently target and degrade the contaminants.[104] Table 2 provides an overview of studies that employed BMOFs for CIP removal.

4.3 Sulfamethoxazole removal

SMX, a widely used antimicrobial, treats infections and supports livestock growth, with global consumption exceeding 84 240 tons annually.[114] Unfortunately, only a small portion of SMX is metabolized or absorbed by living organisms, with approximately 70% being excreted through feces or urine and subsequently discharged into water. However, due to the limitations of current wastewater treatment technologies in effectively removing such antibiotics, significant concentrations of SMX have been detected in the effluent from medical industries, municipal sewage systems, and livestock farms.[115] These SMX residues not only contribute to bacterial resistance and reduce the efficacy of drug treatments but also pose risks to ecosystems and human health.[116] Therefore, it is crucial to develop more effective treatment methods to eliminate SMX.

Tang *et al.* synthesized Fe/Cu-MOF and evaluated its performance in the catalytic degradation of SMX. The BMOF system demonstrated high efficiency for SMX degradation across a broad pH range (4.0–8.6). At an initial pH of 5.6, the BMOF catalyst achieved complete removal of SMX (20 mg L^{-1}) within 120 minutes, outperforming monometallic Fe-MOF and Cu-MOF catalysts.[117] In addition, Wu *et al.* utilized Mn/Fe-MOFs as a cathode in a heterogeneous electro-Fenton system to effectively remove SMX. At pH 3 and a current of 30 mA, the system achieved 96% SMX degradation within 90 minutes, with 12.09 mg L^{-1} of H_2O_2 and 0.21 mM of ·OH detected, highlighting its efficiency.[118] Similarly, Zhou *et al.* developed a novel Fe/Co-MOF for SMX removal in an AOP. The Fe/Co-MOF demonstrated excellent catalytic performance in activating peracetic acid (PAA) for SMX degradation under neutral conditions. While increasing PAA concentration improved SMX removal, varying the Fe/Co-MOF dosage from 0.05 to 0.2 g L^{-1} had minimal impact on degradation efficiency.[119] Furthermore, Xie *et al.* introduced a self-assembly strategy to synthesize highly dispersed Co/Fe bimetallic carbon cages ($CoFe_{50}$@C) through the thermal transformation of Fe-doped dual MOFs. Leveraging the well-dispersed Co/Fe species, synergistic effects, and enhanced carbon graphitization, $CoFe_{50}$@C achieved 98% SMX removal within 180 minutes.[120] Guo *et al.* proposed a dual-MOF-assisted strategy to construct core–shell magnetic Fe_3O_4@ZIFs composites for PAA activation. The Fe_3O_4@ZIFs exhibited superior activity, achieving 99.3% SMX degradation within 30 minutes, outperforming similar materials.[121] Lastly, Peng *et al.* synthesized a stable Fe/Co-MOF to activate peroxymonosulfate (PMS) for SMX degradation. Fe/Co-MOF demonstrated exceptional catalytic performance, achieving 100% degradation of 5 mg per L SMX within 30 minutes.[122]

4.4 Carbamazepine removal

CBZ is a commonly used pharmaceutical compound found in drugs and PPCPs.[123] It is a significant micropollutant due to its widespread use and high detection rate in natural water sources. After CBZ is administered to humans, various derivatives are formed through *in vivo* metabolism and environmental degradation of the parent compound. These derivatives are often more toxic and harder to degrade than CBZ itself, making it essential to study their environmental behavior and develop effective removal methods. Widely used in the treatment of epilepsy and bipolar disorder, CBZ has a high annual consumption rate.[124] Research indicates that prolonged exposure to CBZ can have toxic effects on the central nervous and digestive systems, impair embryonic cell development, and affect blood cell levels.[125] As a result, there is an urgent need to develop effective treatment technologies to eliminate CBZ and its derivatives from aquatic environments.

Several studies have explored efficient catalysts for CBZ degradation. Zheng *et al.* successfully developed a highly efficient Mn-doped MIL-53 (Fe) precursor at high temperatures. The FeMn@C-800/2 catalyst demonstrated the highest catalytic performance for CBZ degradation, achieving an apparent first-order reaction rate 8.9 and 17.8 times greater than Fe@C-800 and Mn@C-800, respectively, under optimal conditions (catalyst dosage: 50 mg L^{-1}, pH: 4.0).[126] Roy *et al.* synthesized NH_2-MIL-125(Ti)@MIL-53(Fe/Co) (AMIL@MIL). This catalyst facilitated CBZ mineralization in aqueous solution *via* PMS activation under visible light, completely degrading CBZ (10 mg L^{-1}) within one hour using 0.05 g L^{-1} of the composite containing 10 wt% NH_2-MIL-125(Ti) and 0.25 g L^{-1} of PMS.[127] Thai *et al.* introduced MIL-100@ZIF-67@MXene, a novel metallic MOF composite anchored on MXene nanosheets, designed for enhanced CBZ degradation and PMS activation. Their study thoroughly investigated the composite's efficiency, reaction parameters, and mechanisms, revealing that the MIL-100@ZIF-67@MXene/PMS system reduced CBZ by 95% within 30 minutes under neutral pH conditions.[128] Huang *et al.* developed Co/N–PC-*T* precursors using solvent heating and immersion methods, followed by simple pot calcination of Co/Zn-MOF to obtain Co/N–PC-*T*. These catalysts were employed for PMS activation and pollutant degradation, with Co/N–PC-800 exhibiting exceptional catalytic performance. When used for PMS activation, Co/N–PC-800 achieved over 98% CBZ degradation in 30 minutes.[129]

4.5 Acetaminophen removal

ACP a pharmaceutical and PPCP, is one of the most widely used painkillers and has been detected in sewage, sewers treatment plants, and even drinking water due to its extensive use.[130] It is also commonly utilized as an analgesic and antipyretic and serves as a key component in anti-flu medications worldwide.[131] ACP in water systems poses significant risks to aquatic life and human health. Research highlights its environmental impact and associated health hazards, including liver failure, gastrointestinal disorders, and liver necrosis.[132] Consequently, there is an urgent need to develop effective methods to remove ACP from wastewater before it is released into aquatic environments.

Alrefaee *et al.* effectively removed pharmaceutical contaminants from wastewater using a novel adsorbent, La/Th-MOF. This material consists of stacked nanorods of 2-methyl imidazole coordinated with lanthanum and thorium. It demonstrated an impressive maximum adsorption capacity of 339.75 mg g^{-1} for ACT, highlighting its potential as a cost-effective and efficient adsorbent for wastewater treatment. The study found that pH levels significantly influence ACT adsorption, with optimal performance occurring in an acidic environment (pH 5) at an adsorbent dosage of 0.02 g.[133] Pattappan *et al.* synthesized Fe/Co-MOF, which exhibited enhanced light absorption in the visible spectrum and a bandgap energy of 1.73 eV. Photoluminescence analysis revealed a lower charge carrier recombination rate in Fe/Co-MOF compared to bare Fe- or Co-MOFs. The Fe/Co-MOF achieved a maximum AAP conversion rate of 97.4% (rate constant 0.031 min^{-1}) in 180 minutes, outperforming Fe-MOF (66%) and Co-MOF (73%). A scavenger study identified superoxide anion radicals as the primary agents responsible for AAP and 2,4-D degradation. The catalyst maintained its stability over five recycles without any decline in AAP degradation efficiency. Fe/Co-MOF photodegraded 2,4-D by 79.8%.[134] Li *et al.* successfully synthesized Fe/Co-MOF by co-doping MIL-101(Fe). This material achieved complete (100%) APAP degradation within 15 minutes at a Fe/Co-MOF concentration of 0.05 g L^{-1} and a PMS concentration of 0.8 mmol L^{-1}. Notably, the degradation process remained effective across a wide pH range (3–9), demonstrating the material's versatility in various wastewater treatment conditions.[135]

4.6 Chloramphenicol removal

CAP is a widely used antibiotic for treating bacterial infections and is frequently detected in surface water, wastewater effluents, groundwater, and soil environments.[136,137] CAP is known for its blood toxicity, embryotoxicity, and potent immunosuppressive effects, which can also disrupt the physiological functions of plants, animals, and microorganisms.[138] As a result, there is an urgent need to develop effective technologies and strategies to eliminate CAP.

Xue *et al.* developed a BMOF derivative to *in situ* modify bulk CA (Ce/Fe@C-CA), creating a bifunctional composite cathode for CAP degradation in the heterogeneous EF process. This composite cathode demonstrated high CAP degradation efficiency of 94.89% was achieved.[139] Lei *et al.* synthesized a novel nitrogen-doped Fe/Ni-MOF derivative for efficient ionizing radiation-catalytic degradation of CAP. Compared to single electron beam (EB) irradiation, the radiation-catalytic process enhanced the degradation rate constant by 2.8 times and improved the total organic carbon (TOC) removal rate by 21.2 times. Notably, a synergistic effect between Fe and Ni in their valence states was observed, with Fe^{2+} playing a crucial role in promoting hydroxyl radical production during the radiation-catalytic process.[140]

4.7 Ibuprofen removal

IBP is a widely used non-steroidal anti-inflammatory drug commonly prescribed for pain relief, fever reduction, and inflammation management.[141] It has proven highly effective in treating rheumatoid arthritis. Due to its extensive global consumption, IBP is frequently detected in freshwater sources, raising concerns about its potential ecological effects. Studies suggest it may have long-term adverse impacts on aquatic life.[142,143] Additionally, ibuprofen has been identified as an indicator of wastewater contamination.[144] Given its threat to aquatic organisms and the stability of ecosystems, it is crucial to remove this pollutant from wastewater before it is released into the environment.

Li *et al.* successfully synthesized Mn-MIL-53(Fe) by adjusting the Mn doping ratio. Using the UV/Mn-MIL-53(Fe)/PMS process, IBP removal reached 79.7% within 30 minutes at a Mn-to-Fe molar ratio of 1.0, with a reaction rate constant 26.9% higher than the undoped counterpart.[145] Similarly, Thai *et al.* developed an advanced bimetallic catalyst, Mn/ZIF-67@GO, for efficient IBP degradation. The Mn/ZIF-67@GO/PMS system exhibited outstanding catalytic performance, achieving 98% degradation of a 0.05 mM IBP solution in just 15 minutes. Additionally, the system proved versatile, successfully removing over 80% of other tested antibiotics.[146]

4.8 Diclofenac sodium removal

DCF, a widely used anti-inflammatory drug, is consumed globally in large quantities. Its high water solubility and polarity contribute to its frequent detection in wastewater, natural water sources, and even drinking water.[147] Prolonged exposure to DCF poses potential health risks, including hemodynamic changes and thyroid tumors.[148] As a result, increasing attention has been directed toward the removal of DCF from aqueous solutions.

Wang *et al.* successfully developed a novel adsorbent based on a Ni/Co-MOF. Kinetic and isothermal analyses revealed that

Table 3 Removal of pharmaceutical residues using BMOFs

Types of BMOFs	Name of residues	Method	Catalyst dosage	Initial concentration	Performance%	Ref.
Fe/Cu-MOF	CAZ	Degradation	—	5 mg L^{-1}	99.5	151
Fe/Ni-MOF	IMB	Adsorption, degradation	99 mg, 50 mg	81 mg L^{-1}, 50 mg L^{-1}	89.12, 92.17	152
Co/Fe-MOF	DOX, 5-FU	Adsorption	0.5 g L^{-1}	10 mg L^{-1}	87.97	153
Co/Cu-MOF	DOX	Degradation	5 mg	20 mg L^{-1}	80	154
Fe/Ni-MOF	ENR	Degradation	2 mg	30 mg L^{-1}	95	155
Fe/Cu-MOF	ENR	Degradation	20 mg L^{-1}	20 mg L^{-1}	90	156
Fe/Cu-MOF	NOR	Degradation	—	20 mg L^{-1}	99.48	157
Co/Zn-MOF	NOR	Adsorption	0.8 g L^{-1}	50 mg L^{-1}	—	158
NI/Mo-MOF	MOX	Degradation		2 mg L^{-1}	95	159
Ni/Co-MOF	CP	Degradation	0.25 g L^{-1}	46.5 mg L^{-1}	88.9	160
Co/Ni-MOF	Sertraline	Degradation	75 mg	117 mg L	97.19	161
Fe/Mn-MOF	OFX	Degradation	0.1 g L^{-1}	5 mg L^{-1}	81.85	162
Fu/Cu-MOF	OFX	Degradation	30 mg	30 mg L^{-1}	100	163
Zr/Co-MOF	Cefradine	Adsorption	2 mg	20 mg L^{-1}	95	164

the adsorption behavior of Ni/Co-BTC MOF for DCF closely followed the Langmuir and pseudo-second-order models, with a maximum adsorption capacity of 343.05 mg g^{-1}.[149] Similarly, He *et al.* synthesized an environmentally friendly Bi–Zr bimetallic MOF derived from plant-based materials and investigated its adsorption properties for typical pharmaceutical and PPCPs, specifically DCF.[150]

4.9 Removal of other pharmaceutical residues

BMOFs have been used to remove various pharmaceutical residues, as summarized in Table 3, including ceftazidime (CAZ), imatinib (IMB), doxorubicin (DOX), 5-fluorouracil (5-FU), enrofloxacin (ENR), norfloxacin (NOR), moxifloxacin (MOX), cefoperazone (CP), sertraline, ofloxacin (OFX), and cefradine. Table 3 presents a summary of studies that employed BMOFs for various pollutant removal.

5 Conclusion and prospect

The presence and behavior of pharmaceuticals in the environment, particularly in aquatic systems, have been a significant focus of scientific research over the past two decades. Due to their biologically active, lipophilic nature and resistance to biodegradation, pharmaceuticals can accumulate and persist in the environment, posing risks even at low concentrations. To address this issue, BMOFs have emerged as promising materials for removing pharmaceutical residues. These materials function as both adsorbents and catalysts, offering unique advantages such as high surface area, exceptional porosity, customizable pore sizes, and structural tunability. Compared to monometallic compounds, BMOFs exhibit enhanced electrical conductivity, greater charge capacity, increased active sites, and adjustable chemical reactivity. While BMOFs demonstrate excellent adsorption and degradation capabilities, challenges remain in achieving consistent pore structures, durable designs, and stable functional groups. Furthermore, ecotoxicological analyses and life-cycle assessments are essential for evaluating the environmental impact of BMOFs, particularly in large-scale applications. Despite these challenges, BMOFs and their composites hold significant potential as advanced materials for pharmaceutical removal, contributing to sustainable environmental practices.

Data availability

Data sharing is not applicable to this article as no datasets were generated or analysed during the current study.

Conflicts of interest

There are no conflicts to declare.

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