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# Heavy metal pollution in aquatic environments and removal using highly efficient bimetallic metal–organic framework adsorbents

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Rapid urbanization and industrial development worldwide have significantly increased the release and spread of anthropogenic heavy metals, extending their impact from local sources to broader regions. This growing pollution poses serious risks to human health and aquatic ecosystems. Although extensive research has been conducted on the removal of heavy metal from water, existing treatment methods still require optimization for improved efficiency and specificity. Among these, adsorption is recognized as the most effective technique, with bimetallic metal–organic frameworks (BMOFs) emerging as highly promising adsorbents due to their exceptional adsorption capabilities and potential to address complex environmental challenges. Therefore, it is essential to implement measures that reduce heavy metal concentrations in water to safe levels. This review provides a detailed account of the sources and toxicity of heavy metals to humans and ecosystems. It explains BMOFs, their synthesis, and mechanisms of interaction with heavy metals, and, for the first time, explores the application of BMOFs for the removal of heavy metals from aqueous environments. In summary, this review offers a comprehensive overview of the latest progress in BMOF-based heavy metal remediation, providing valuable insights for future BMOF synthesis and practical solutions for water decontamination.

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## 1. Introduction

The availability of clean, high-quality water is essential for social, human health, and economic development, and the stability of global ecosystems.<sup>1–3</sup> As water is a fundamental need for all forms of life, rapid urban growth and industrialization have significantly increased its demand. Beyond daily human consumption, water plays a vital role in numerous sectors, including residential use, agriculture, petroleum refining, pharmaceuticals, and medical applications. However, these activities often introduce harmful pollutants and waste materials into water sources.<sup>4</sup> Many industrial processes, in particular, generate hazardous waste that poses a serious threat to environmental and public health.

Heavy metal pollution, in particular, has become one of the most pressing environmental challenges.<sup>5</sup> With the rapid expansion of industries such as mining, metal plating, fertilizer production, tanneries, battery manufacturing, pesticide use and paper production, the release of heavy metals into the environment is on the rise, particularly in developing countries. In contrast to organic pollutants, heavy metals are non-biodegradable and can build up in living organisms. Many of these metals, such as zinc, copper, cadmium, nickel, mercury,

chromium, and lead, are known to be toxic or carcinogenic.<sup>6</sup> The effective removal of heavy metals from water systems remains a critical yet challenging task for environmental engineers. Heavy metal contamination in terrestrial and aquatic ecosystems is a major environmental issue with serious public health implications. Although many heavy metals occur naturally, anthropogenic activities have significantly increased their concentrations.<sup>7</sup> Removal is essential due to the wide-ranging hazards they pose: to humans, they can cause liver damage, respiratory, neurological, and cardiovascular disorders; to ecosystems, they impair plant growth, reduce biodiversity, and disrupt the behavioral, biochemical, physiological, and reproductive functions of aquatic organisms; to water sources, they degrade quality, alter physical and chemical properties, disturb ecological balance, and reduce dissolved oxygen; and to industries, they result in regulatory noncompliance, costly remediation, and reputational damage.<sup>2,8–10</sup>

Several techniques have been developed for the removal of heavy metals, including adsorption,<sup>11</sup> ion exchange,<sup>12</sup> membrane filtration,<sup>13</sup> chemical precipitation,<sup>14</sup> and electrochemical methods.<sup>15</sup> Among these, adsorption stands out due to its design flexibility, operational simplicity, and ability to produce high-quality treated water. Moreover, many adsorbents can be regenerated through cost-effective and efficient desorption processes, allowing for repeated use.<sup>16,17</sup> As a result, adsorption has emerged as a leading method for the removal of heavy metals from contaminated water and wastewater.

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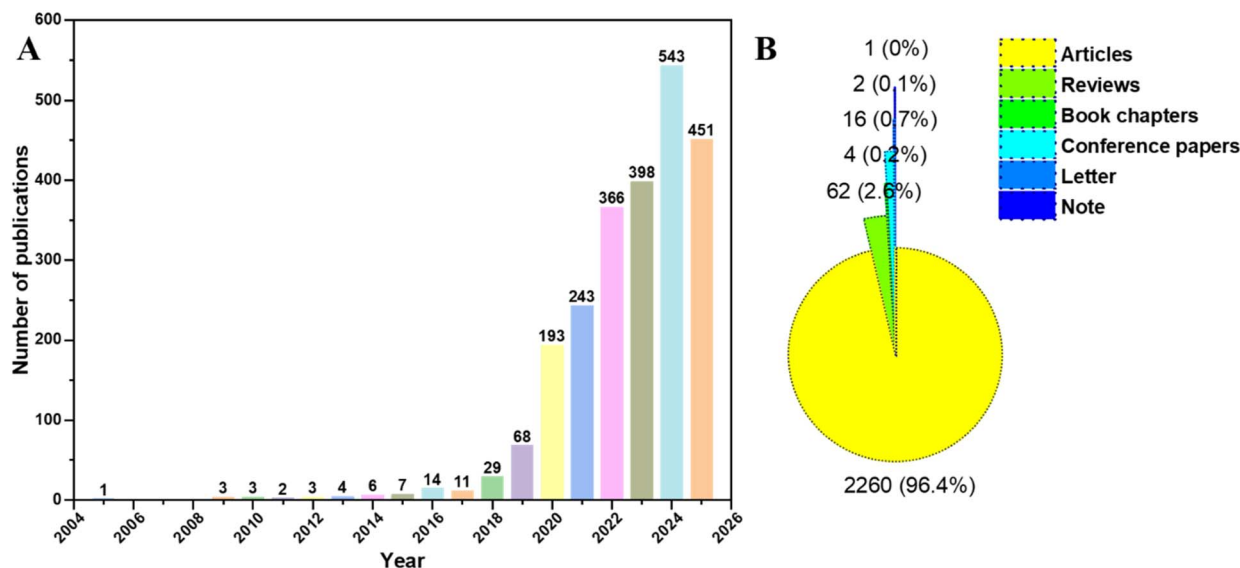


Fig. 1 (A) Number of papers published on "Metal–Organic Frameworks" and "Bimetallics," and (B) types of papers published from 2005 to 2025, based on a Scopus database search.

Although numerous researchers have employed various adsorbents such as activated carbon, zeolites, lignite coke, ash, activated alumina, clay, and natural fibers to remove heavy metal ions from wastewater, the adsorption capacity and selectivity largely depend on the chemical and physical characteristics of both the adsorbent and the adsorbate.<sup>18</sup> Consequently, over the past decade, increasing attention has been given to developing more effective and advanced adsorbents through the combination of different composite materials.

Metal–organic frameworks (MOFs) represent a unique class of porous materials formed by metal ions coordinated with organic ligands, resulting in one-, two-, or three-dimensional structures.<sup>19–21</sup> Numerous studies have identified MOFs as highly promising materials in diverse fields such as catalysis,<sup>22</sup> separation,<sup>23</sup> sensing,<sup>24</sup> drug delivery,<sup>25</sup> water treatment,<sup>26–32</sup> and energy storage and conversion.<sup>33</sup> Their exceptional properties including adjustable pore size, high surface area, tunable structures, and versatile chemical composition make them especially suitable for adsorption-based applications.<sup>34,35</sup> In the field of water treatment, MOFs have demonstrated outstanding potential due to their excellent porosity and surface functionality.<sup>36</sup> However, some limitations remain, including low water stability, limited chemical resistance, and the presence of micropores that can restrict access to certain target ions.<sup>37,38</sup> To overcome these challenges, researchers have explored the development of BMOFs by incorporating two different metal ions into the framework. This integration enhances both the physical and chemical properties, often resulting in a synergistic effect or dual-function mechanism that improves overall stability and performance.<sup>39–43</sup> Therefore, BMOFs, synthesized by incorporating different metal ions, often demonstrate superior adsorption performance for heavy metal removal compared to their monometallic counterparts.

This review stands out for its exclusive focus on the application of BMOFs as adsorbents for heavy metal removal an area

that has not been specifically addressed in previous reviews. A search of the Scopus database using the keywords "Metal–Organic Frameworks" and "Bimetallics" revealed significant growth in related publications between 2005 and 2025. This upward trend is illustrated in Fig. 1A, which shows a sharp increase in cumulative publications, particularly over the past five years, indicating a growing interest in BMOFs for various applications. Additionally, the distribution of publication types is presented in Fig. 1B. All data were retrieved from the Scopus database in July 2025.

Numerous research groups have investigated various approaches for treating wastewater contaminated with heavy metals using different materials.<sup>31–33</sup> This review summarizes the application of BMOFs for heavy metal removal, highlighting their superior adsorption performance compared to other materials. It also examines the occurrence of heavy metals, their adverse effects on the environment and human health, the introduction of BMOFs, the interaction mechanisms between BMOFs and heavy metals, and the potential future applications of BMOFs in this field. To the best of our knowledge, no prior review has thoroughly addressed the use of BMOFs specifically for heavy metal removal. Thus, this paper seeks to present current insights into the application of BMOFs for treating wastewater contaminated with toxic metals and to propose future research directions. As illustrated in Fig. 2.

### 1.1 Scientific novelty of this review

Numerous reviews have been published on MOFs across environmental fields, often focusing on quality assessments in the existing literature. For example, Adil *et al.* summarized the use of MOF-based nanofibers for heavy metal removal,<sup>44</sup> Mubarak *et al.* reviewed MOFs for removing heavy metals from contaminated water,<sup>45</sup> and Yuan *et al.* provided an overview of heavy metal removal from water *via* adsorption on MOFs.<sup>46</sup>

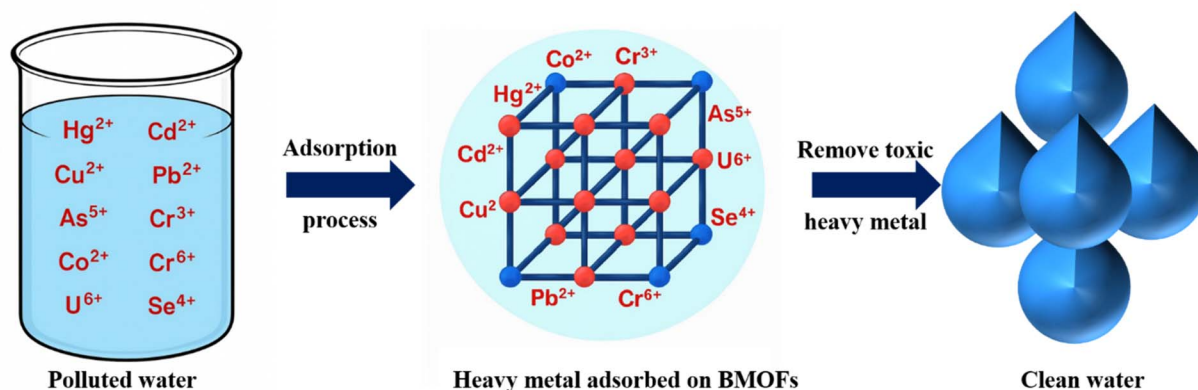


Fig. 2 Schematic diagram illustrating the removal of heavy metals by BMOFs.

Although previous studies have made significant contributions, no comprehensive review has specifically focused on BMOFs for heavy metal removal. Recent research on the use of BMOFs in various applications has garnered increasing attention. This review, for the first time, examines the emerging field of BMOFs, providing a detailed overview of their potential in environmental remediation. It discusses the sources and toxicity of heavy metals to human health and ecosystems, the characteristics and synthesis methods of BMOFs, and the adsorption mechanisms between heavy metals and BMOFs. Furthermore, the review highlights the application of BMOFs in removing metals such as chromium, mercury, uranium, copper, lead, and other metals. These insights contribute to a deeper understanding of BMOFs as innovative adsorbents for heavy metal removal, emphasizing the significance and timeliness of this review in the environmental field.

## 2. Heavy metals: sources and toxicity to human health and ecosystems

Heavy metals originate from both natural and anthropogenic sources, each contributing to environmental contamination to varying degrees. Natural sources include volcanic eruptions, rock weathering, forest fires, biological activity, sea spray, and other geochemical processes through which heavy metals are introduced into ecosystems. Natural disasters particularly floods, but also earthquakes, hurricanes, tornadoes, and tsunamis can significantly influence the release and distribution of heavy metals into aquatic environments.<sup>47</sup> While these natural inputs are generally less harmful and can often be assimilated by the environment, anthropogenic sources are far more significant and detrimental. Human activities such as smelting, mining, fossil fuel combustion, industrial effluent discharge, sewage sludge disposal, agricultural runoff, and emissions from vehicles contribute substantially to heavy metal pollution. Atmospheric deposition and rainfall can further transfer these contaminants from air and soil into water bodies.<sup>48</sup> Among these, mining and smelting are the most

dominant contributors, with smelting alone accounting for an estimated 40–73% of total anthropogenic heavy metal emissions.<sup>49,50</sup> Despite their environmental severity, anthropogenic sources are more manageable through regulatory measures, technological advancements, and improved industrial practices. Therefore, controlling human-derived pollution is critical for reducing heavy metal contamination and protecting ecological and human health.<sup>51</sup>

The primary contributors to heavy metal pollution in the environment include activities such as milling, mining, metal plating, and surface finishing industries.<sup>52</sup> These sectors are major sources of a wide range of toxic metals released into the atmosphere, including chromium (Cr), cobalt (Co), zinc (Zn), copper (Cu), lead (Pb), cadmium (Cd), and nickel (Ni).<sup>53</sup> Over the past several decades, the concentrations of these metals in water bodies and sediments have significantly increased. Consequently, the accumulation of heavy metals in agricultural soils has led to elevated levels of toxic metals in grains and vegetables, posing a serious threat to both human health and the environment. This risk is exacerbated by the toxic, non-biodegradable, and bioaccumulative nature of heavy metals, which enables them to persist in ecosystems and enter the food chain.<sup>54</sup> Exposure to these metals can lead to a range of adverse health effects, including carcinogenicity, neurotoxicity, hepatotoxicity, nephrotoxicity, and disruption of endocrine and immune system functions.<sup>55</sup>

In ecosystems, heavy metals disrupt biological processes and alter the structure and function of microbial, plant, and animal communities.<sup>49</sup> In soils, they can reduce microbial diversity, inhibit enzymatic activity, and suppress plant growth by interfering with nutrient uptake.<sup>56,57</sup> In aquatic environments, heavy metals can bioaccumulate in fish and other organisms, leading to oxidative stress, reproductive impairments, and increased mortality.<sup>58</sup> These effects can cascade through food webs, ultimately threatening biodiversity and ecosystem stability.<sup>59</sup> Due to their persistence and cumulative toxicity, heavy metals are classified as priority pollutants by environmental protection agencies worldwide.<sup>8</sup> Therefore, controlling their release and



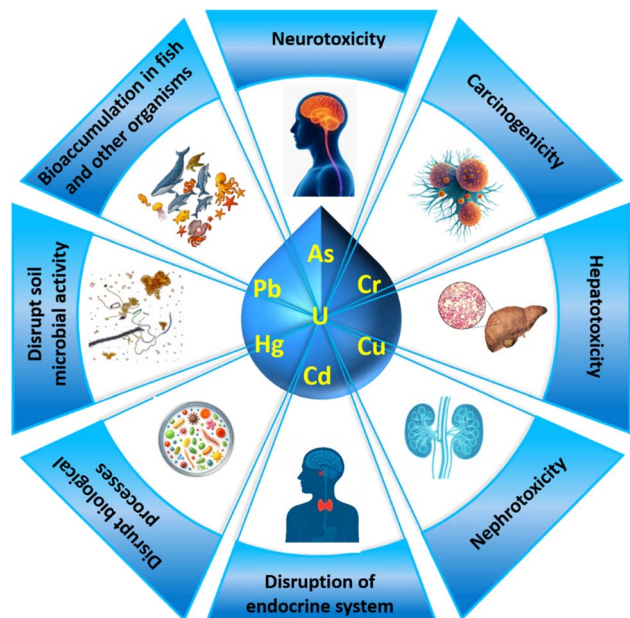


Fig. 3 Toxicity of heavy metals for human health and ecosystems.

mitigating their impacts through effective remediation strategies and policy interventions is critical to protecting both environmental and public health. The toxicity of heavy metals for both humans and the environment is depicted in Fig. 3.

### 3. Bimetallic metal–organic frameworks advantages

Traditionally, a variety of adsorbents such as carbon-based materials, minerals, macromolecules, and biomass have been employed for the removal of heavy metal ions from aqueous media. However, these conventional adsorbents suffer from limitations such as poor selectivity and relatively slow adsorption kinetics, often attributed to disordered pore structures that hinder the effective transport of metal ions.<sup>60</sup> Additionally, pretreatment steps such as acidification or functionalization with specific chemical groups are often required to enhance adsorption capacity and selectivity due to the weak coordination interactions between the adsorbent and metal ions.<sup>61</sup> Recently, MOFs have emerged as highly promising materials for the removal of heavy metals from contaminated water, owing to their exceptional adsorption efficiency.<sup>62</sup> MOFs are crystalline porous materials constructed from metal ions coordinated with organic linkers, forming three-dimensional hybrid networks.<sup>63,64</sup> Their high surface area, tunable porosity, and well-defined pore structures make them particularly suitable for water purification applications.<sup>65,66</sup>

To further improve their performance, recent research has focused on developing bimetallic MOFs by incorporating two different metal ions into the framework.<sup>67</sup> This approach introduces synergistic effects by enabling partial substitution at the metal nodes or secondary building units (SBUs), enhancing structural stability and allowing the tuning of physicochemical

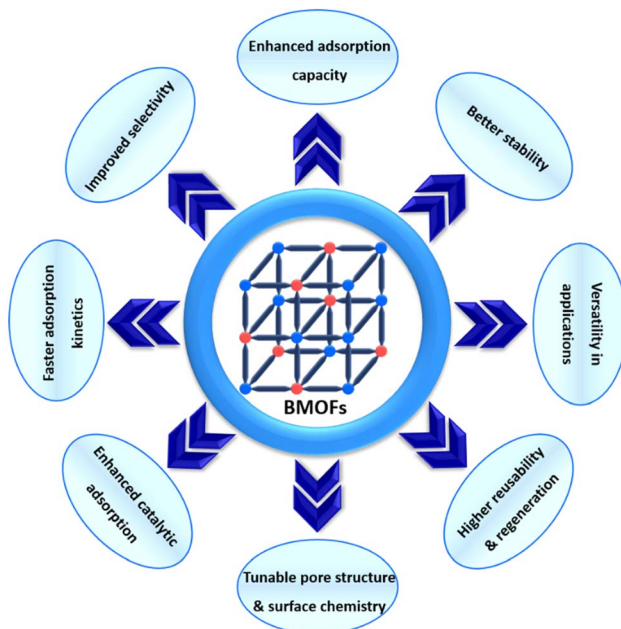


Fig. 4 Comparison of the key advantages of BMOFs over conventional adsorbent.

properties.<sup>68</sup> Bimetallic MOFs show superior performance compared to monometallic counterparts, particularly in catalysis, sensing, gas storage, and drug delivery.<sup>69</sup> In environmental remediation, their enhanced surface characteristics, increased number of active sites, and stronger host–guest interactions contribute to a significantly higher maximum adsorption capacity ( $q_m$ ) for a wide range of heavy metal ions. For example, a Ni/Cd-MOF demonstrated a  $q_m$  of 950.61 mg g<sup>-1</sup> for Pb(II), approximately twice that of the single-metal Ni-MOF.<sup>70</sup> Similarly, the adsorption performance for arsenic was notably improved in the Fe/Al-BDC-NH<sub>2</sub> bimetallic MOF synthesized by Yin *et al.*, which incorporated both iron and aluminum, compared to the corresponding monometallic frameworks Fe-BDC-NH<sub>2</sub> and Al-BDC-NH<sub>2</sub>.<sup>71</sup> These findings suggest that BMOFs, prepared by incorporating different metal elements, can exhibit superior adsorption properties compared to their monometallic counterparts. The advantages of BMOFs over other adsorbents are illustrated in Fig. 4.

#### 3.1 Synthesis of bimetallic metal–organic frameworks

As shown in Fig. 5, it presents a schematic of two synthesis routes for BMOFs: (a) one-step *in situ* synthesis and (b) post-synthetic modification. In the one-step approach, two different metals and organic linkers are combined simultaneously to form a BMOF. The post-synthetic method involves a two-step process, in which an MOF is first prepared using conventional methods, followed by the partial replacement of primary metal atoms with secondary ones to enhance electrical conductivity through the formation of hetero-surfaces.<sup>72</sup>

BMOFs can be synthesized *via* several approaches. One-pot synthesis (OPS) integrates two metal salts in a single reaction, forming complex networks with synergistic effects, while

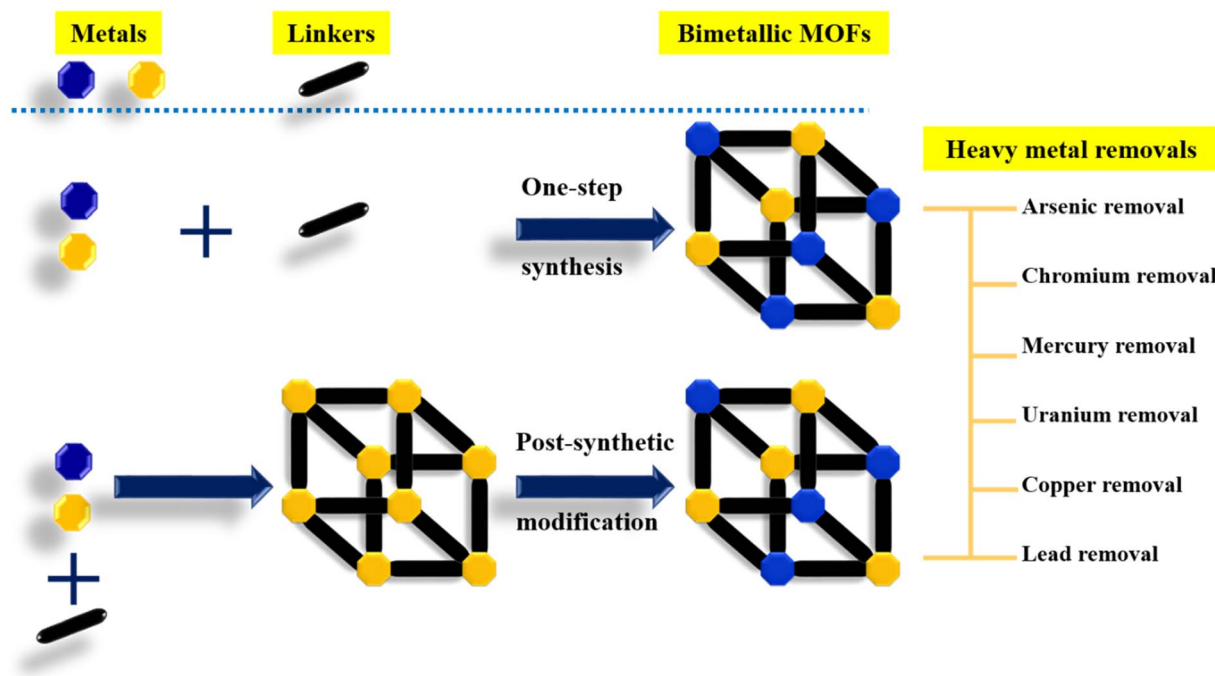


Fig. 5 Schematic of BMOF synthesis via one-step and post-synthetic approaches.

simplifying the process and reducing intermediate steps.<sup>73</sup> Post-synthetic modification uses metal ion exchange in preformed MOFs, influenced by factors such as coordination number, ionic radius, lattice flexibility, and solvent; for example,  $\text{Cu}^{2+}$  often replaces  $\text{Cd}^{2+}$ ,  $\text{Zn}^{2+}$ , or  $\text{Mn}^{2+}$ , whereas  $\text{Cd}^{2+}$  and  $\text{Pb}^{2+}$  exchange faster due to labile bonds.<sup>68</sup> Direct synthesis combines metal ions during MOF formation but can yield brittle frameworks with unpredictable properties, requiring careful control of pH, reactivity, solubility, and coordination to achieve the desired metal ratio.<sup>69</sup> Template methods regulate metal composition and can produce hollow BMOFs with enhanced active sites and mass transport, using either self-template (dissolution-regrowth) or exterior-template (sacrificial template removal) strategies.<sup>41</sup>

### 3.2 Forces between bimetallic metal-organic framework adsorbents and heavy metal ion adsorbates

The efficiency of heavy metal removal by BMOFs depends on the interaction forces between the adsorbent and the adsorbate. These forces are typically classified into two categories: chemical adsorption (chemisorption) and physical adsorption (physisorption).<sup>74,75</sup> Physisorption, or physical adsorption, takes place when adsorbate particles attach to the adsorbent surface *via* weak intermolecular forces, such as van der Waals interactions. In contrast, chemisorption refers to the process where adsorbate molecules attach to the adsorbent surface through chemical forces or the formation of chemical bonds.<sup>76</sup> MOFs exhibit enhanced adsorption capabilities due to the synergistic effects arising from the incorporation of two different metal ions within their structures. Primarily, coordination bonds form between the unsaturated metal sites in the MOF and heavy metal ions, facilitated by electron pair donation from donor

atoms (e.g., oxygen, nitrogen) present in organic linkers or metal nodes. The presence of two distinct metal centers expands the diversity and density of these active sites, allowing stronger and more selective binding to heavy metal ions.<sup>77</sup> For example, Yu *et al.* prepared a novel Zn/Ni-MOF. The  $\text{Hg(II)}$  adsorption capacity of the BMOF reached  $744.4 \text{ mg g}^{-1}$ . More impressively, the BMOF successfully removed 99.99% of mercury. This suggests that the main adsorption mechanism of the BMOF is driven by the strong coordination between the  $-\text{SH}$  groups and mercury ions, with electrostatic attraction playing a lesser role.<sup>78</sup>

Electrostatic interactions also play a critical role, particularly when bimetallic MOFs possess tunable surface charges that attract oppositely charged metal ions, thereby enhancing adsorption. For instance, Zeng *et al.* successfully synthesized a bimetallic coordination polymer (Ti/Zr-TA) for the removal of mercury from wastewater. The material exhibited high adsorption capacities of 583.5, 615.4, and  $654.4 \text{ mg g}^{-1}$  at different temperatures. The study further revealed that the adsorption mechanism involved both electrostatic interactions and chelation, with the latter specifically the coordination between mercury ions and oxygen- or sulfur-containing functional groups playing the dominant role.<sup>79</sup> In addition, ion-exchange mechanisms are significant, as metal ions within the MOF framework can be replaced by heavy metal ions in solution, thereby facilitating efficient capture. For example, the Zn/Cu-BTC- $\text{NH}_2$  metal-organic framework (MOF) enhances  $\text{Pb}^{2+}$  uptake by promoting amino group-mediated adsorption and enabling ion exchange between copper and lead ions. The enrichment mechanism was identified as a combination of  $\text{Pb}^{2+}$  adsorption through  $-\text{NH}_2$  functional groups and ion exchange involving the substitution of Cu ions with  $\text{Pb}^{2+}$ .<sup>80</sup> Lastly, during



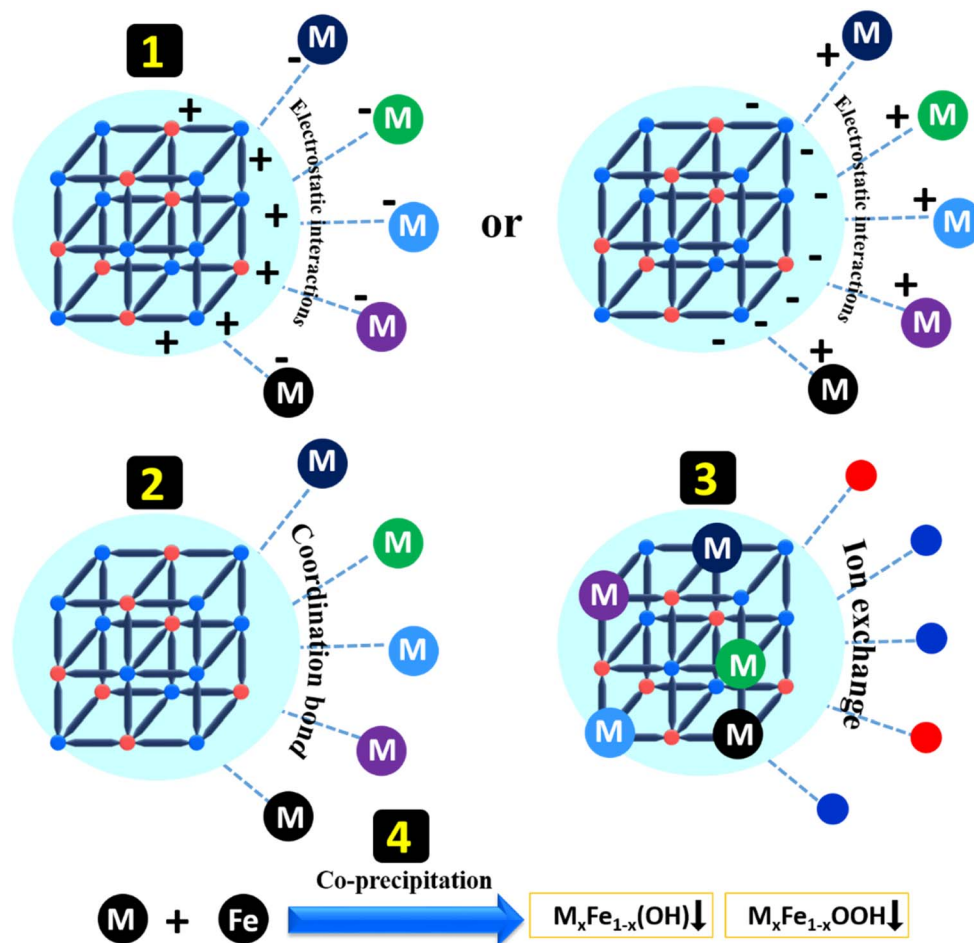


Fig. 6 Schematic illustration of the mechanism between heavy metals and BMOFs.

co-precipitation, contaminants become physically or chemically bound within the iron oxides generated by the corrosion of zero-valent iron, resulting in their immobilization and subsequent removal from water.<sup>81</sup> Collectively, these forces provide bimetallic MOFs with superior adsorption capacity, selectivity, and stability compared to their monometallic counterparts, making them highly effective for heavy metal remediation (Fig. 6).

## 4. Removal of heavy metals from aquatic environments

BMOFs that have been studied for the removal of heavy metal ions from water are described. Table 1 summarizes all the studies discussed, providing an overview of the key information reported in the literature. The following subsections are divided by the specific metal adsorbed and discuss the findings of each study in detail, based on the information provided by the respective authors.

### 4.1 Arsenic removal

Arsenic is a ubiquitous metalloid element that primarily exists in nature in the form of various compounds. These arsenic

compounds can be found in sediments, soil, and water, and have been classified as Group 1 carcinogens due to their high toxicity. Ecotoxicological studies have reported that excessive exposure to arsenic and long-term consumption of arsenic-contaminated drinking water may lead to various diseases, including cancer, hyperkeratosis, and liver and cardiovascular disorders.<sup>105</sup> Guo *et al.* developed a Fe/Zr-MOFs for the effective decontamination of arsenic in water. The adsorption capacities of BMOF for arsenate [As(v)] and arsenite [As(III)] reached 204.1 mg g<sup>-1</sup> and 101.7 mg g<sup>-1</sup>, respectively. Additionally, the material effectively removed arsenic from real water samples (1.0 mg L<sup>-1</sup>) within 2 hours, achieving removal efficiencies of 99.0% for As(III) and 99.8% for As(v).<sup>82</sup> In addition, Fang *et al.* synthesized magnetic Zn/Co-MOFs incorporating both catalytic and adsorption functionalities. These MOFs were combined with PMS to enable the simultaneous oxidation of As(III) and adsorption of the resulting As(v). The PMS notably enhanced the initial adsorption rates rising from 31.19 to 358.96 μg g<sup>-1</sup> min<sup>-1</sup> for As(III), and from 354.12 to 497.85 μg g<sup>-1</sup> min<sup>-1</sup> for As(v).<sup>83</sup>

Furthermore, Han *et al.* successfully synthesized bimetallic La-Zr MOFs through a simple fabrication method. The resulting material was applied as an adsorbent for the removal of



**Table 1** An overview of the key information on the BMOFs discussed in this review. The Table is sorted according to metal ions, followed by the chemical formula of the BMOFs used, adsorption capacity, pH, temperature, performance

| Metal                     | BMOF                          | Adsorption capacity (mg g <sup>-1</sup> )       | Optimal pH | Temperature (K) | Performance % | Ref. |
|---------------------------|-------------------------------|---|------------|-----------------|---------------|------|
| As                        | Fe/Al-BDC-NH <sub>2</sub>     | 146.8   | 5–9        | 298             | —             | 71   |
| As (III,V)                | UiO-66(Fe/Zr)                 | 101.7, 204.1                                    | 7          | 298             | 99, 99.8      | 82   |
| As(III,V)                 | Zn/Co-MOF                     | 1.242, 2.063                                    | 5          | 298             | —             | 83   |
| As(III,V)                 | Fe/Mn-MOF                     | 344.14, 228.79                                  | 11         | —               | 77.99, 99.59  | 84   |
| As(v), Cr                 | La/Zr-MOF                     | 83.40, 222.5                                    | 2          | 298             | —             | 85   |
| As(III)                   | Co/Mn MOF                     | 531   | 11         | —               | 93.4          | 86   |
| As(III)                   | Fe/Mn-MOF                     | 152.5   | 7          | —               | —             | 87   |
| Hg                        | Ti/Zr-TA                      | 583.5, 615.4 and 654.4                          | 5          | 298–318         | —             | 79   |
| Hg                        | Cu/Ni-BTC                     | 144.32  | 5          | 298             | —             | 88   |
| Hg                        | Zn/Ni-MOF                     | 744.4   | 4          | —               | 99.99         | 78   |
| U                         | Cu/Fe-BTC                     | 354   | 7          | —               | —             | 89   |
| U                         | Ce/Mn-MOF                     | 1218.78   | 6.5        | 313             | 88.6          | 90   |
| U                         | Mg/Mn-MOF                     | 113.9   | 4          | 298             | 93.88         | 91   |
| Cu                        | Ni/Co-MOF                     | 233.99  | 5          | 298             | —             | 92   |
| Cd, Cu                    | Ag/Fe-MOF                     | 265, 213  | 5          | 298             | —             | 93   |
| Cr                        | Cu@MIL-53(Fe)                 | 724.6   | 3          | 298             | 99.05         | 94   |
| Cr(vi)                    | Ce-UiO-66-NH <sub>2</sub> @LS | 282.77  | 2          | —               | 94.90         | 95   |
| Cr                        | Fe/Zr-MOF                     | 75.55   | 5          | 303             | 91.56         | 96   |
| Cr                        | (Fe/Co)-BDC                   | 588.23  | 5.3        | —               | —             | 97   |
| Cr                        | Zr/Ce-UiO-67                  | —   | 1          | —               | 93            | 98   |
| Cr                        | Fe/Cu-BDC                     | —   | 3          | —               | 98            | 99   |
| Cu, Cr, U                 | Ni/Co-MOF                     | 756.82  | 5.6        | —               | —             | 18   |
|                           |                               | 111.22  |            |                 |               |      |
|                           |                               | 365.25  |            |                 |               |      |
| Pb                        | Fe doped HKUST-1              | 565   | 5          | —               | 90            | 100  |
| Pb                        | Zr/Ce MOF                     | 667.04  | 6          | 298             | —             | 101  |
| Co                        | Co/Ni-MOF                     | 372.66  | 6          | —               | —             | 102  |
| Pb, Cd                    | Co/Fe-MOF                     | 193.4, 182.2                                    | 5,7        | 298             | —             | 103  |
| Pb(II), Cu(II) and Cd(II) | Fe/Mg-MOF                     | 140.6, 95.56, 134.4                             | 5,7        | —               | —             | 39   |
| Se(IV), (VI)              | UiO-66(Fe/Zr)                 | 196 mg g <sup>-1</sup> , 258 mg g <sup>-1</sup> | 5          | 298–318         | —             | 104  |

Cr(vi) and As(v). The maximum monolayer adsorption capacity for H<sub>3</sub>AsO<sub>4</sub> was 83.40 mg g<sup>-1</sup>, primarily due to ligand exchange and coordination interactions involving La–OH and Zr–OH groups. Moreover, the sample labeled <sub>1</sub>LaUN<sub>12</sub> demonstrated excellent Cr(vi) adsorption performance, achieving a maximum monolayer capacity of 222.5 mg g<sup>-1</sup>. Notably, over 40% of the adsorbed Cr(vi) was reduced to Cr(III) by amino functional groups and subsequently immobilized on the surface of <sub>1</sub>LaUN<sub>12</sub>.<sup>85</sup> Additionally, Yang *et al.* synthesized Fe/Mn bimetallic MOF materials, which demonstrated outstanding adsorption performance for As(v) and As(III), with maximum theoretical *q<sub>m</sub>* of 228.79 mg g<sup>-1</sup>, and 344.14 mg g<sup>-1</sup>, respectively.<sup>84</sup> Lastly, Zhang *et al.* designed an FeMn-MOF-74 adsorbent that provided combined adsorption and oxidation sites, creating a synergistic effect for the removal of As(III). It achieved the highest experimental *q<sub>m</sub>* (161.6 mg g<sup>-1</sup>) reported among MOF-based arsenic adsorbents.<sup>87</sup>

## 4.2 Chromium removal

Chromium is extensively used in many manufacturing industries. Its compounds typically occur in two oxidation states: trivalent (Cr(III)) and hexavalent (Cr(VI)). Cr(VI) ions are highly soluble in the form of oxyanions, whereas Cr(III) ions tend to precipitate as Cr(OH)<sub>3</sub> and exhibit lower solubility.<sup>106,107</sup> The hexavalent form is regarded as more toxic and hazardous than

the trivalent form because of its greater stability and carcinogenic nature.<sup>108</sup> Continuous accumulation of Cr<sup>6+</sup> along the food chain may pose serious health risks, such as kidney damage, and gastric, dermatitis, lung cancer, eye and respiratory tract irritation, and can lead to biomagnification.<sup>109</sup> Yuan *et al.* successfully synthesized a novel Cu/Fe-MOF material and applied it for Cr(vi) removal. The Cr(vi) removal *q<sub>m</sub>* were 20.65 mg g<sup>-1</sup> at 180 minutes and 13.35 mg g<sup>-1</sup> at 15 minutes. At 25 °C, 45.55% of total chromium and 99.05% of Cr(vi) were removed, with a *q<sub>m</sub>* of 724.6 mg g<sup>-1</sup>.<sup>94</sup> Similarly, Koppula *et al.* impregnated MnO<sub>2</sub> particles into a NiCo(BDC) BMOF to synthesize a highly stable NiCo(BDC)@MnO<sub>2</sub> composite. This material was used for the removal of Cu(II), Cr(vi), and U(vi) metal ions. The NiCo(BDC)@MnO<sub>2</sub> composite demonstrated high *q<sub>m</sub>* of 756.82 mg g<sup>-1</sup> for Cu(II), 365.25 mg g<sup>-1</sup> for U(vi), and 111.22 mg g<sup>-1</sup> for Cr(vi).<sup>18</sup>

In another study, Wang *et al.* fabricated a Ce-UiO-66-NH<sub>2</sub>@LS composite that proved effective for the removal of both methyl orange and Cr(vi). Under acidic conditions, the *q<sub>m</sub>* for Cr(vi) reached 282.77 mg g<sup>-1</sup>, with a removal efficiency of 94.90%. The material maintained over 80% removal efficiency after four consecutive reuse cycles.<sup>95</sup> Furthermore, Zhao *et al.* synthesized Fe/Zr-MOFs for the effective removal of Cr(vi) ions from wastewater. Optimal removal performance was achieved at pH 5, with an initial Cr(vi) concentration of 20 mg L<sup>-1</sup> at 303 K.



The Fe/Zr-MOFs exhibited a removal efficiency of up to 82% even after five consecutive reuse cycles, demonstrating their stability and reusability in repeated adsorption processes.<sup>96</sup> Abuzalat *et al.* synthesized (Fe/Co)-BDC using a simple solvothermal method. Its anion exchange adsorption capacity was evaluated against chromium oxyanions, which are recognized as hazardous pollutants. The material exhibited a remarkable  $q_m$  of 588 mg g<sup>-1</sup> for Cr-oxyanions.<sup>97</sup> Lastly, Yu *et al.* synthesized a bimetallic UiO-66(Zr/Al) material, UZA-6, which demonstrated outstanding photocatalytic performance by removing 99.25% of hexavalent chromium (Cr(vi)) from water. Furthermore, the catalyst efficiently adsorbed the resulting trivalent chromium (Cr(III)), sustaining a removal efficiency of 97.73% and thereby ensuring the complete elimination of chromium from the solution.<sup>110</sup>

#### 4.3 Mercury removal

Mercury is a highly toxic metal that bioaccumulates in living organisms and poses significant risks to human health. It primarily originates from industrial and household sources.<sup>111,112</sup> Methylmercury, in particular, can lead to neurological, genetic, pulmonary, and renal disorders, especially in children.<sup>113</sup> Sokhansanj *et al.* synthesized CuNi-BTC@Fe<sub>3</sub>O<sub>4</sub> for mercury removal, achieving a  $q_m$  of 144.32 mg g<sup>-1</sup>, which fit well with the equilibrium data.<sup>88</sup> In addition, Zhang *et al.* synthesized FeCu-MOFs enriched with chlorine and applied them for the removal of elemental mercury (Hg<sup>0</sup>). The findings indicated that the FeCu-MOFs exhibited good crystallinity and uniform elemental distribution. These bimetallic MOFs achieved high Hg<sup>0</sup> removal efficiencies, exceeding 90% under various conditions. The presence of O<sub>2</sub>, HCl, and NO in the flue gas further enhanced the mercury removal efficiency. In addition, the FeCu-MOFs demonstrated strong resistance to water vapor and SO<sub>2</sub> poisoning. Adsorption kinetics analysis showed that the equilibrium mercury  $q_m$  reached 12.27 mg g<sup>-1</sup> at 393 K.<sup>114</sup>

#### 4.4 Uranium removal

Uranium pollution has become a growing concern with the rapid expansion of nuclear power. When uranium enters the environment, it poses significant ecological risks. As a result, uranium-contaminated wastewater requires effective treatment.<sup>115</sup> At the same time, recovering uranium from wastewater would be highly beneficial to the nuclear industry. Uranium, characterized by its radioactivity, chemical toxicity, and non-degradability, poses significant risks to both human health and the ecological environment.<sup>116</sup> Liu *et al.* designed and synthesized two MOF materials, Cu-BTC and Cu/Fe-BTC, for the adsorption of U(vi). The optimum pH for Cu-BTC was 3.0, and at this pH, the  $q_m$  for U(vi), was 617 mg g<sup>-1</sup>. For Cu/Fe-BTC, the optimal pH was 7.0, with a  $q_m$  of 354 mg g<sup>-1</sup> at this pH.<sup>89</sup> Similarly, Li *et al.* developed a novel bimetallic Ce/Mn-modified aminated MOF, which demonstrated excellent performance as an adsorbent for radioactive decontamination. The material exhibited a maximum U(vi)  $q_m$  of 1218.78 mg g<sup>-1</sup> at 313 K. Notably, after five adsorption-desorption cycles, the removal efficiency remained above 83.5%.<sup>90</sup> Lastly, Hassanin *et al.*

developed bimetallic Mg/Mn-based MOFs for uranium adsorption. The highest  $q_m$ , approximately 113.90 mg g<sup>-1</sup>, was achieved at pH 4.0 after 60 minutes.<sup>91</sup>

#### 4.5 Copper removal

Copper pollution originates from various industrial activities, including mining, metal plating, fiber production, and electronics manufacturing, as well as from agricultural practices and contaminated food or packaging materials. Exposure to copper poses significant health risks, including neurotoxicity (e.g., Wilson's disease), kidney damage, liver disorders, and a potentially increased risk of lung cancer due to prolonged inhalation of copper-containing sprays.<sup>117,118</sup> Kong *et al.* developed MOFs-CMC composites, including Ni/Co-MOF, Ni-MOF, and Co-MOF. The adsorption behavior of these composites toward Cu<sup>2+</sup> was investigated through batch adsorption experiments, adsorption kinetics, and isotherm studies. The  $q_m$  followed the order: Co-MOF (214.38 mg g<sup>-1</sup>), Ni/Co-MOF (233.99 mg g<sup>-1</sup>), and Ni-MOF (216.95 mg g<sup>-1</sup>), indicating a synergistic effect between Ni and Co that enhanced Cu<sup>2+</sup> adsorption.<sup>92</sup> Similarly, El-Yazeed *et al.* successfully synthesized a Ag/Fe-MOF, which showed outstanding  $q_m$  for Cd(II) and Cu(II), reaching 265 mg g<sup>-1</sup> and 213 mg g<sup>-1</sup>, respectively. Among the tested MOFs, the Ag-Fe (0.6 : 1) framework exhibited superior adsorption performance.<sup>93</sup>

#### 4.6 Lead removal

Lead is one of the most acutely toxic heavy metals. Even very low concentrations of lead ions in drinking water can cause diseases such as hepatitis, encephalopathy, high blood pressure, miscarriage, kidney damage, and impaired brain and central nervous system function in unborn children when it crosses the placenta.<sup>119,120</sup> Goyal *et al.* investigated the impact of iron doping on the hydrostability and adsorption behavior of HKUST-1 MOFs. Due to their high surface area, microporosity, and coordinatively unsaturated metal sites, the Fe-doped HKUST-1 exhibited remarkable selectivity and adsorption capacity for Pb(II), achieving over 90% removal efficiency and a  $q_m$  of 565 mg g<sup>-1</sup>.<sup>100</sup> Additionally, Dahui An *et al.* investigated Zr/Ce MOF for the removal of Pb(II) from aqueous solutions, achieving a  $q_m$  of 667.04 mg g<sup>-1</sup> at pH 6 and 298 K.<sup>101</sup> Lastly, Li *et al.* investigated both Ni-MOF and Ni/Cd-MOF materials. The bimetallic Ni/Cd-MOF, compared to the monometallic Ni-MOF, featured fewer layers, a larger specific surface area, greater pore size, and higher surface electronegativity. These properties contributed to its outstanding performance in Pb<sup>2+</sup> removal, with a significantly higher adsorption rate, stronger affinity, and a superior  $q_m$  of 950.61 mg g<sup>-1</sup> nearly double that of Ni-MOF.<sup>70</sup>

#### 4.7 Other heavy metal removal

In addition to the aforementioned metals, the removal of other heavy metals such as cadmium (Cd), cobalt (Co), selenium (Se), and various combinations of multiple heavy metals has also been extensively investigated. The key findings including removal efficiencies, conditions, and BMOF types are comprehensively summarized in Table 1. Alshorifi *et al.* developed (Fe/

Co) bimetallic MOF catalysts that effectively removed toxic metal ions ( $\text{Hg}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Cd}^{2+}$ , and  $\text{Cu}^{2+}$ ) from aqueous solutions, exhibiting high adsorption capacities. The maximum removal efficiencies were 95.98% for lead, 98.89% for mercury, 93.89% for cadmium, and 92.6% for copper ions.<sup>121</sup> In addition, Sengupta *et al.* developed Zr/Zn bimetallic MOFs, with Zr–Zn–MOF-1 showing the  $q_m$  of 77.4% within 3 hours at pH 5. They also investigated how pH influenced the surface charge of the MOFs to better understand the relationship between pH and sorption performance. The  $q_m$  reached 129.0 mg g<sup>−1</sup>.<sup>122</sup> Furthermore, Mazlan *et al.* studied rGO-PDA/Co-ZIF-8 aerogels, which demonstrated an exceptionally high  $q_m$  of 1217 mg g<sup>−1</sup> for lead, with a removal efficiency exceeding 99%. The material also exhibited excellent adsorption performance for other heavy metals, achieving  $q_m$  of 1163 mg g<sup>−1</sup> for copper and 1059 mg g<sup>−1</sup> for cadmium. Notably, the lead adsorption capacity remained stable at 1023 mg g<sup>−1</sup> with a removal efficiency above 80% even after seven cycles at pH 6.<sup>123</sup> Lastly, Shen *et al.* reported that the adsorption capacities of CdK-*m*-COTTB reached 636.94 mg g<sup>−1</sup> for  $\text{Pb}^{2+}$ , 432.90 mg g<sup>−1</sup> for  $\text{Tb}^{3+}$ , and 357.14 mg g<sup>−1</sup> for  $\text{Zr}^{4+}$  ions.<sup>124</sup>

## 5. Challenges and future perspectives

It highlights the unique structural and chemical properties of BMOFs, such as tunable porosity, large surface areas, and synergistic effects of dual metals, which enhance adsorption performance for heavy metal removal. The review elucidates the mechanisms of heavy metal adsorption, including physisorption, chemisorption (such as coordination bonds, electrostatic interactions, ion-exchange mechanisms, and coprecipitation). It identifies design strategies for BMOFs, including the choice of metal combinations, ligands, and synthesis methods, to optimize their performance for removing different metals.

By summarizing the latest advances and challenges, the review opens avenues for developing multifunctional, cost-effective, and scalable BMOFs for environmental remediation. While previous studies have explored monometallic MOFs for heavy metal removal, no comprehensive review has systematically addressed the unique advantages of BMOFs for this purpose. Many prior studies focus on individual case studies or single-metal frameworks, whereas this review integrates these findings, identifies knowledge gaps, and provides a holistic perspective on the potential of BMOFs for heavy metal remediation. Therefore, the insights provided here are novel and not fully captured in the earlier literature.

Despite the promising performance of BMOFs in removing toxic heavy metals from aquatic environments, several challenges remain that must be addressed to advance their practical application. One major limitation is the stability of BMOFs in aqueous and chemically harsh environments, where structural degradation can hinder both adsorption efficiency and recyclability. Additionally, large-scale synthesis of BMOFs with consistent quality, controlled composition, and uniform porosity presents both technical and economic challenges. Issues such as potential leaching of metal ions, high production

costs, and limited regeneration cycles further restrict their implementation in real-world wastewater treatment systems.

Moreover, the adsorption mechanisms at the molecular level are not yet fully understood, particularly the synergistic interactions between the two metal centers and their roles in the selective binding of heavy metal ions. Comprehensive studies integrating advanced characterization techniques and theoretical modeling are needed to elucidate these complex interactions. Environmental compatibility, toxicity assessments, and long-term performance evaluations are also critical for ensuring the safe deployment of BMOFs in natural ecosystems.

Looking ahead, future research should prioritize improving the chemical and structural stability of BMOFs under diverse environmental conditions, developing green and cost-effective synthesis routes, and enhancing selectivity and reusability through surface modification or functionalization. The integration of BMOFs into composite materials, membranes, or hybrid systems may also pave the way for scalable and multifunctional water purification technologies. Furthermore, interdisciplinary collaboration spanning materials science, environmental engineering, and computational chemistry will be essential for accelerating innovation and translating laboratory-scale advances into industrial-scale solutions for sustainable heavy metal remediation.

## 6. Conclusion

BMOFs have emerged as highly efficient and versatile adsorbents for the removal of toxic heavy metals from aqueous environments. By integrating two different metal nodes, BMOFs exhibit enhanced structural stability, tunable porosity, abundant active sites, and synergistic effects that significantly improve adsorption capacity and selectivity compared to their monometallic counterparts. Heavy metal removal occurs primarily through physisorption or chemisorption interactions between metal ions and the active sites within the BMOF structure.

Despite their remarkable potential, challenges remain in scaling up production, improving long-term stability under harsh environmental conditions, and ensuring cost-effective regeneration. Future research should focus on developing eco-friendly synthesis methods, exploring novel metal combinations, and integrating BMOFs into continuous treatment systems to bridge the gap between laboratory studies and large-scale applications. Overall, BMOFs represent a promising next-generation platform for sustainable and high-performance heavy metal adsorption and environmental remediation.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

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



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