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Review

Mild Interfacial Catalysis for Sustainable Water Remediation: Active-Site Regulation, Non-Radical Oxidation, and Ecological Compatibility

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Abstract

Sustainable water remediation requires catalytic strategies that remove contaminants efficiently while reducing chemical input, byproduct formation, and ecological disturbance. Conventional radical-dominated advanced oxidation processes can rapidly degrade pollutants, but their reliance on high oxidant dosages and freely diffusing reactive oxygen species often causes matrix quenching, non-selective oxidation, low oxidant utilization, and potential ecological risks. Mild interfacial catalysis provides a materials-chemistry strategy to regulate oxidative intensity and direct contaminant transformation under environmentally relevant conditions. In this review, mild catalysts are defined by pathway-selective, interfacially confined, and environmentally compatible oxidation rather than by low dosage alone. Representative non-radical or low-intensity pathways, including singlet oxygen generation, surface-mediated electron transfer, high-valent metal–oxo species, and direct oxidative transfer processes, are discussed in relation to active-site structure, oxidant utilization, matrix tolerance, and byproduct control. We further summarize how coordination environments, defect chemistry, heteroatom configurations, nanoconfinement, and immobilized interfaces regulate reactive-species formation and interfacial charge transfer. Key material platforms, including single-atom catalysts, heteroatom-doped carbons, defect-engineered oxides, catalytic membranes, hydrogels, and floating or immobilized composites, are evaluated from mechanistic and application-oriented perspectives. Finally, catalyst regeneration, cost, microbial community responses, algae–bacteria balance, ecotoxicity, and long-term safety are discussed to guide sustainable aquatic ecosystem restoration.

Keywords: mild interfacial catalysis; sustainable water remediation; non-radical oxidation; active-site regulation; catalyst immobilization; ecological compatibility

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1. Introduction

Against the backdrop of increasing pressure on water quality and the growing prominence of emerging contaminants, achieving pollution control with lower chemical inputs and reduced ecological disturbance has become an important issue at the interface of water environmental management, environmental materials science, and sustainable

catalysis [1–3]. Current challenges, including black-odorous rivers and lakes, internal loading from sediments, accumulation of trace organic contaminants, and residual pollutants in reclaimed water, are often not caused by excessive concentrations of a single contaminant alone. Rather, they arise from the combined effects of long-term low-dose exposure, complex matrix interactions, and imbalanced ecological processes [4–6]. Although conventional "high-intensity removal" strategies can rapidly decrease target contaminant concentrations, their application in real environmental systems is often accompanied by high reagent consumption, poor reaction selectivity, increased byproduct risks, and substantial disturbance to in situ ecological processes [7,8]. Therefore, shifting from "stronger oxidation" toward more sustainable chemical transformation has become an important direction in the development of water remediation technologies [9].

Advanced oxidation processes (AOPs) have played an important role in advanced wastewater treatment and the removal of recalcitrant organic contaminants because of their strong oxidative capacity [10,11]. However, conventional radical-dominated systems usually rely on relatively high oxidant dosages and highly reactive species to maintain removal efficiency, raising increasing concerns regarding their process sustainability. On the one hand, although hydroxyl radicals and sulfate radicals possess extremely strong, non-selective oxidative capacities, they are readily quenched or transformed by background constituents such as carbonate, natural organic matter, and halide ions in complex water matrices, thereby reducing oxidant utilization efficiency and increasing operational costs [12,13]. On the other hand, strong radical pathways may induce non-directed transformation of organic contaminants, generating intermediates that are more difficult to identify and control and thereby increasing the risks of byproduct toxicity and environmental safety concerns [8]. In addition, some conventional oxidation systems require stringent pH conditions, external energy input, or strict control of metal leaching, which limits their suitability for sustainable, low-maintenance, and in situ deployment [14]. Accordingly, relying solely on high-intensity radical oxidation is not fully consistent with the principles of green chemistry, which emphasize reducing hazardous chemical inputs, improving reaction efficiency, minimizing byproduct formation, and enhancing process safety.

In this context, mild catalysis has attracted growing attention as a chemical remediation strategy that places greater emphasis on reaction controllability and process sustainability [15–17]. Unlike conventional intensified oxidation, mild catalysis does not aim to maximize instantaneous oxidative strength. Instead, it relies on the rational design of catalytic materials to preferentially construct non-radical or low-intensity oxidative pathways under lower oxidant dosages and milder reaction conditions, thereby enabling selective transformation of target contaminants and risk reduction [18,19]. Such systems typically depend on specific catalytic interfaces to induce active processes such as singlet oxygen generation, surface-mediated electron transfer, or high-valent metal–oxo species formation, improving contaminant removal while reducing unnecessary oxidant consumption and byproduct formation risks [20].

In this review, a "mild catalyst" is not defined simply by a low catalyst dosage or a low pollutant removal rate [21,22]. Rather, it refers to a catalytic material that is intentionally designed or operated to moderate oxidative intensity and guide oxidant activation toward pathway-selective, interfacially confined, and environmentally compatible transformations [23,24]. More specifically, a mild catalyst should preferentially satisfy several criteria: (i) it activates oxidants through non-radical or low-intensity interfacial pathways, such as singlet oxygen, surface-mediated electron transfer, high-valent metal–oxo species, or direct oxidative transfer, rather than relying predominantly on freely diffusing •OH or SO_4^{-} [25,26]; (ii) it achieves effective contaminant transformation at lower oxidant input by improving oxidant utilization efficiency and reducing bulk-phase radi-

cal loss; (iii) it exhibits higher selectivity toward target pollutants or key toxic functional groups, thereby lowering the probability of uncontrolled oxidation and harmful byproduct formation [27,28]; and (iv) it maintains structural stability, low leaching, recoverability, or immobilized operation under environmentally relevant water conditions [29].

Accordingly, the boundary between a mild catalyst and a conventional AOP catalyst should be drawn mainly according to the dominant reaction pathway and the intended function of catalyst design, rather than according to catalyst composition alone. Conventional AOP catalysts are typically designed to maximize the generation of highly reactive radical species and rapid pollutant degradation or mineralization, often at the expense of oxidant efficiency, matrix tolerance, and byproduct control. In contrast, mild catalysts are designed to regulate where, how, and to what extent oxidative equivalents are delivered to pollutants, with emphasis on interfacial reaction control, selective transformation, reduced secondary risks, and compatibility with downstream ecological or biological processes. This distinction is not always absolute, because the same catalytic material may behave as a conventional radical-generating AOP catalyst under one set of conditions but as a mild catalyst under another, depending on oxidant dosage, pH, water matrix, surface state, and reactor configuration. Therefore, "mildness" should be understood as a reaction–material–operation attribute, not merely as an intrinsic label of a material.

More importantly, this concept is highly consistent with the design logic of functional materials for sustainable water remediation: lowering chemical input by improving oxidant utilization efficiency, reducing secondary pollution through pathway-selective catalysis, extending process lifetime through stable and reusable material architectures, and enabling in situ or low-energy operation through immobilized catalytic systems [30,31]. As summarized in Figure 1, the conceptual shift from conventional high-intensity oxidation to mild catalysis involves not only lowering oxidant dosage and reducing byproduct formation, but also redesigning catalytic materials, active-site structures, and interfacial reaction pathways to achieve selective transformation, ecological compatibility, and long-term aquatic ecosystem restoration.

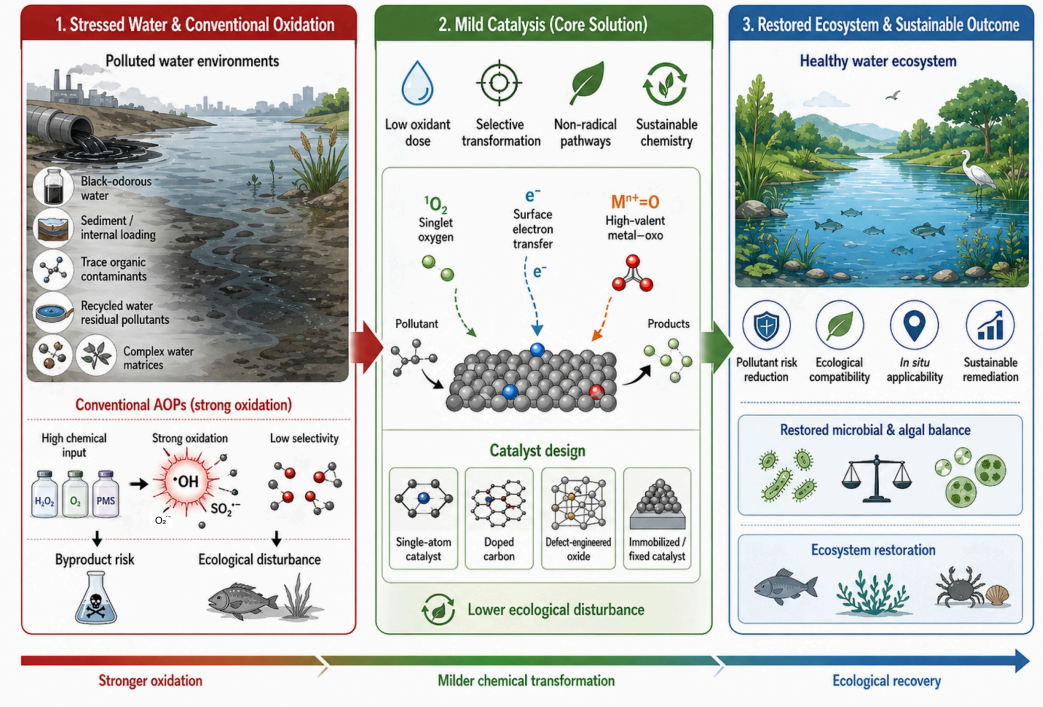


Figure 1. Conceptual framework for the transition from high-intensity oxidation to mild catalysis for sustainable water remediation.

From a materials perspective, the realization of mild catalysis does not depend simply on a particular oxidant, but more fundamentally on the structure–property–function relationships of catalytic materials. In recent years, the development of defect-engineered metal oxides, heteroatom-doped carbon materials, single-atom catalysts, and immobilized composite catalytic systems has enabled researchers to regulate oxidant activation from the perspectives of coordination environment, electronic structure, interfacial chemistry, and confinement effects. These advances make it possible to guide reactions away from conventional strong radical-dominated pathways toward more selective and environmentally compatible mild oxidation processes [32–35]. This indicates that the key question in water remediation is no longer merely whether contaminants can be degraded or mineralized, but whether effective transformation, organic-carbon fate regulation, and catalyst regeneration can be achieved through sustainable catalytic material design at the lowest possible resource and environmental cost [36]. Re-examining mild catalysis from the perspective of materials chemistry and sustainable catalysis can therefore help explain differences among catalytic systems in reaction efficiency and byproduct behavior, while also establishing material design principles for practical applications.

This concept is particularly important for aquatic ecosystem restoration. Scenarios such as black-odorous water treatment, pollution control at the sediment–water interface, and micropollutant removal require technologies that balance chemical effectiveness, ecological compatibility, and engineering feasibility [37–39]. Excessively aggressive oxidation may improve physicochemical indicators in the short term, but it can also impose additional disturbances on microbial community structure, algae–bacteria interactions, and benthic ecological processes. In contrast, mild catalysis may provide a more reasonable balance between pollution control and ecological recovery through low-dose operation, reduced byproduct formation, immobilized architectures, and in situ deployability [40–42]. In other words, the value of mild catalysis lies not only in contaminant removal, but also in reducing systemic risks through more sustainable chemical pathways, preserving ecological recovery potential, and providing a material and process foundation for long-term remediation under real environmental conditions [43].

Several related reviews have summarized advanced oxidation processes for trace organic contaminant removal, oxidative water treatment, persulfate activation, non-radical oxidation mechanisms, single-atom-catalyst-mediated AOPs, catalytic membrane systems, and immobilized or floating photocatalysts [44–48]. These studies have provided important foundations for understanding oxidant activation chemistry, reactive-species identification, catalyst structure–activity relationships, and process engineering [49,50]. However, most existing reviews treat non-radical reaction chemistry, catalytic material design, immobilized architectures, and ecological safety as separate topics [51,52]. In many cases, catalytic systems are still evaluated mainly by degradation efficiency, ROS identification, or material activity, whereas the connections among active-site chemistry, low-dose pathway-selective oxidation, byproduct suppression, catalyst form factor, and ecological compatibility remain insufficiently integrated [53].

The originality of this review lies in positioning mild interfacial catalysis as a materials-chemistry framework for sustainable water remediation, rather than simply as another branch of AOPs. Compared with previous reviews, this work makes four main contributions. First, it defines “mildness” from the perspective of reaction-pathway control, oxidant utilization, byproduct-risk reduction, and environmental compatibility, instead of equating it only with low catalyst loading or high pollutant removal. Second, it links molecular-level active-site regulation, including coordination environment, defect chemistry, heteroatom configuration, nanoconfinement, and interfacial electron transfer, with non-radical and low-intensity oxidative pathways. Third, it extends the discussion from nanoscale cat-

alytic activity to macroscopic material form, including catalytic membranes, hydrogels, filters, and floating or immobilized composites for continuous-flow and in situ deployment. Finally, it incorporates ecological compatibility into the evaluation of catalytic materials by considering catalyst leaching, particle release, transformation-product toxicity, microbial community responses, algae–bacteria balance, and long-term environmental stability. Therefore, this review aims to provide an integrated framework that connects catalytic chemistry, materials design, process applicability, and ecological safety for low-dose and low-disturbance water remediation.

Accordingly, this review summarizes recent progress in mild catalytic materials for aquatic remediation from a materials-design perspective. The discussion focuses on how catalyst composition, coordination structure, defect chemistry, electronic configuration, interfacial environment, nanoconfinement, and immobilized architecture regulate oxidant activation and determine reaction selectivity. First, the chemical basis of non-radical and low-dose oxidation pathways is discussed as a mechanistic foundation for materials design. Second, representative material platforms, including single-atom catalysts, heteroatom-doped carbon materials, defect-engineered metal oxides, catalytic membranes, hydrogels, and floating or immobilized composites, are reviewed in terms of their structure–property–function relationships. Third, the form-factor design of catalytic materials is evaluated for continuous-flow treatment, open-water deployment, sediment-interface control, and micropollutant removal. Finally, challenges related to mass transfer, catalyst leaching, structural aging, biofouling, ecological compatibility, regeneration, cost, and scale-up are discussed. This review aims to provide a materials-focused framework for developing low-dose, low-byproduct, stable, and ecologically compatible catalytic systems for aquatic ecosystem restoration.

2. Mechanisms and Regulation of Non-Radical and Low-Dose Oxidation Pathways

2.1. Radical Versus Non-Radical Oxidation Mechanisms

During advanced oxidation processes, oxidants such as persulfate, peroxymonosulfate, and hydrogen peroxide can be activated through either radical or non-radical pathways, depending on the catalyst structure, oxidant dosage, solution chemistry, and interfacial environment [54]. Radical pathways mainly involve freely diffusing reactive oxygen species, such as hydroxyl radicals (•OH) and sulfate radicals (SO_4•$^-$). These radicals possess high redox potentials and can rapidly attack a wide range of organic contaminants through hydrogen abstraction, electron transfer, and addition reactions [55]. Therefore, radical-dominated AOPs are particularly useful when fast degradation or strong mineralization is required. However, their high reactivity also means low selectivity. In real water matrices, •OH and SO_4•$^-$ can be rapidly consumed by carbonate, bicarbonate, chloride, bromide, natural organic matter, and other background constituents, which lowers oxidant utilization efficiency and increases chemical demand [56,57]. In addition, radical reactions usually occur in the bulk solution or near the catalyst surface without strong molecular recognition, making transformation pathways difficult to control and increasing the possibility of halogenated byproducts or toxic intermediates.

Non-radical mechanisms differ from radical pathways in several key aspects. First, the dominant oxidizing species or processes are usually more surface-confined or substrate-selective, including singlet oxygen ($^1O_2$), surface-mediated electron transfer, high-valent metal–oxo species, and direct oxidative transfer processes [58]. Second, non-radical pathways are generally less dependent on freely diffusing ROS and therefore can reduce ineffective oxidant consumption in the bulk phase. Third, many non-radical processes preferentially oxidize electron-rich moieties or adsorbed pollutants at the catalyst–water interface,

which improves reaction selectivity and matrix tolerance. Fourth, because the reaction is more closely related to pollutant adsorption, catalyst electronic structure, and interfacial charge transfer, the oxidative pathway can be tuned by active-site design, coordination regulation, defect engineering, heteroatom doping, and nanoconfinement. These features make non-radical oxidation particularly attractive for mild catalysis, where the objective is not simply maximum instantaneous oxidation strength, but controlled contaminant transformation with lower oxidant input and reduced ecological disturbance.

The reason this review places emphasis on non-radical and low-intensity interfacial pathways is therefore directly related to the concept of mild catalysis. In aquatic ecosystem restoration, the target is often not complete mineralization at any cost, but selective transformation, toxicity reduction, byproduct suppression, matrix tolerance, and compatibility with downstream biological or ecological processes. Radical-dominated systems remain important benchmarks and may be useful at the initial stage of treating high contaminant loads. However, if the process relies mainly on uncontrolled bulk-phase $\bullet OH/SO_4\bullet^-$ generation, it may conflict with the goals of low-dose, low-byproduct, and low-disturbance remediation. By contrast, non-radical pathways provide a more suitable chemical basis for mild catalysis because they allow oxidative equivalents to be generated and consumed near the catalyst–pollutant interface, thereby improving oxidant utilization and reducing unnecessary reactions with non-target matrix components.

It should also be emphasized that the distinction between radical and non-radical mechanisms is not always absolute. Many catalytic systems show mixed radical and non-radical behavior, and the dominant pathway may shift with pH, oxidant concentration, dissolved oxygen, inorganic ions, natural organic matter, pollutant structure, and catalyst aging. Therefore, identifying a non-radical pathway should not rely on a single quenching experiment alone. Instead, reliable mechanistic assignment should combine scavenging tests, probe reactions, electron paramagnetic resonance, isotope or solvent effects, electrochemical analysis, in situ spectroscopy, product analysis, and matrix-tolerance evaluation. This critical distinction is essential for determining whether a catalytic system can truly be regarded as "mild" rather than merely a low-dosed conventional radical AOP.

As shown in Figure 2a, a metal-free N/F dual-site catalyst can promote the selective generation and nearby utilization of $^1O_2$ from PMS through electronic-structure modulation and nitrogen-vacancy construction. This example illustrates how catalyst design can direct oxidant activation toward a non-radical pathway. Together with the surface-mediated electron-transfer and direct oxidative-transfer processes shown in Figure 2b,c [59,60], these mechanisms provide the chemical foundation for the mild interfacial catalysis framework discussed in this review.

2.2. Representative Non-Radical Reactive Species and Low-Dose Oxidation Pathways

Non-radical pathways commonly involved in mild catalytic systems mainly include singlet oxygen ($^1O_2$), surface-mediated electron transfer, high-valent metal–oxo species, and direct oxidative transfer processes [24,54,58–60]. These mechanisms differ fundamentally from radical-dominated oxidation. Radical pathways rely mainly on freely diffusing $\bullet OH$ and $SO_4\bullet^-$, which possess high oxidative strength and can rapidly degrade a broad range of contaminants, but they are usually short-lived, poorly selective, and readily quenched by natural organic matter, carbonate/bicarbonate, chloride, bromide, and other matrix constituents [54–56]. As a result, radical-dominated systems often require higher oxidant input and may generate uncontrolled transformation products or halogenated byproducts in complex waters.

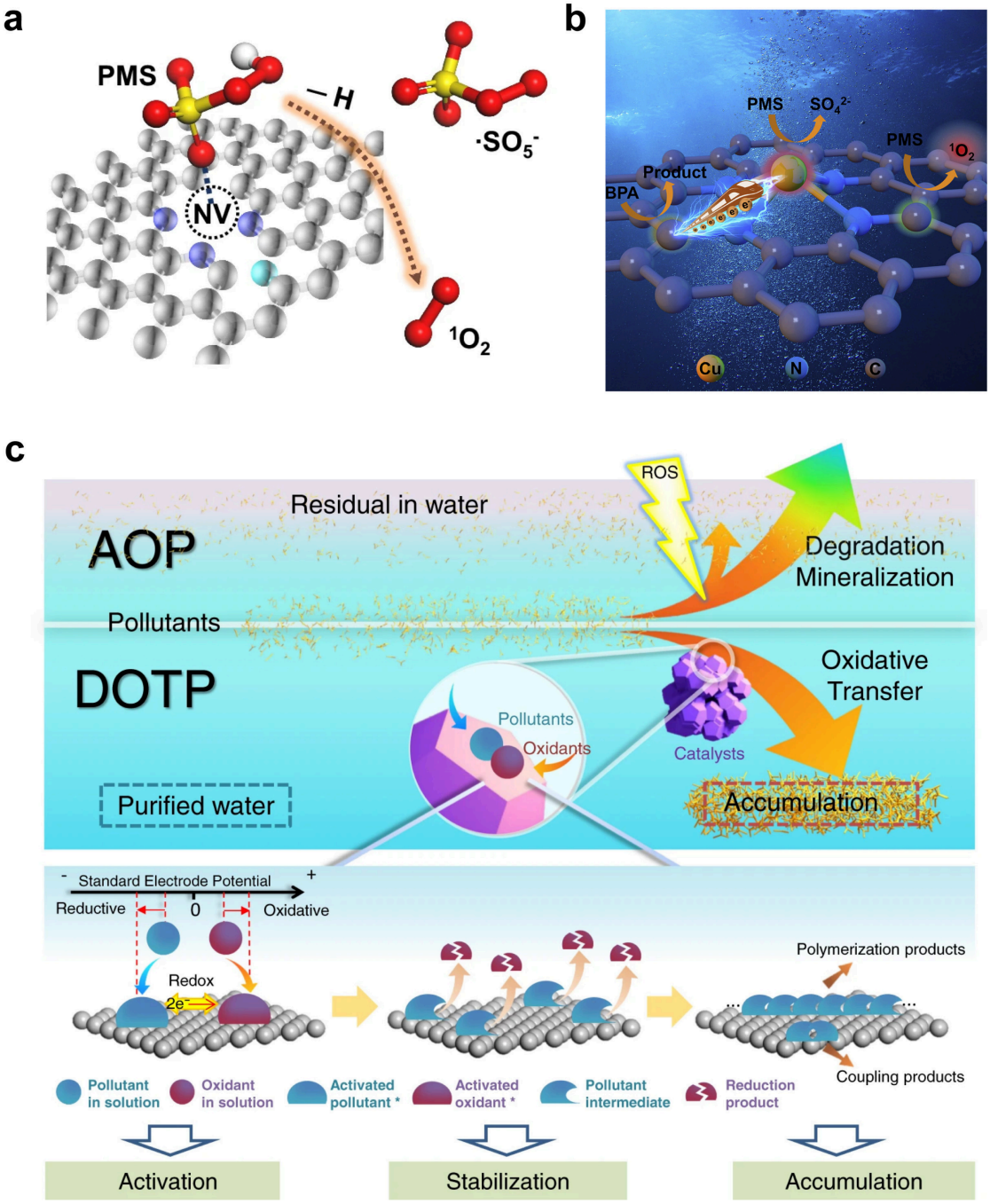


Figure 2. (**a**) Generation and in situ utilization of singlet oxygen ($^1O_2$). Reprinted by permission from ref. [18]. Copyright (2024) Springer Nature. (**b**) Surface-mediated electron-transfer pathway. Reprinted by permission from ref. [59]. Copyright (2022) Elsevier. (**c**) Low-dose surface oxidative transfer process. Reprinted by permission from ref. [60]. Copyright (2022) Springer Nature.

By contrast, non-radical pathways are generally more interfacial, selective, and controllable. $^1O_2$ can selectively oxidize electron-rich moieties and usually shows better resistance to common water-matrix scavengers [56,59]. Surface-mediated electron-transfer processes depend on the adsorption and electronic coupling among pollutants, oxidants, and catalytic interfaces, thereby reducing the ineffective consumption of reactive species in the bulk solution [59,61]. High-valent metal–oxo species, such as Fe(IV)=O or Co(IV)=O, behave more like surface-confined oxidants whose reactivity is regulated by the metal coordination environment, electronic structure, and substrate-binding mode [58,62,63]. Direct oxidative transfer further emphasizes the directional delivery of oxidative equivalents from surface-

activated oxidants to surface-associated pollutants, rather than the accumulation of freely diffusing ROS in solution.

Therefore, the non-radical approach is emphasized in this review because it better matches the central objective of mild catalysis: achieving effective contaminant transformation under lower oxidant input while improving matrix tolerance, reaction selectivity, byproduct control, and ecological compatibility. Radical pathways remain important and may be useful for rapidly reducing high contaminant loads, but a process dominated by uncontrolled bulk-phase •OH/SO_4•$^-$ generation is less consistent with low-dose, low-disturbance aquatic remediation. In this sense, non-radical and interfacially confined pathways provide a more suitable chemical foundation for designing mild catalytic materials. However, the distinction between radical and non-radical mechanisms should be made critically, because many systems involve mixed pathways and their dominant mechanism may change with catalyst structure, oxidant dosage, pH, water matrix, and reaction configuration. Mechanistic identification should therefore combine scavenging experiments, probe reactions, electron paramagnetic resonance, electrochemical analysis, product analysis, and matrix-tolerance tests, rather than relying on a single diagnostic method.

It should be further clarified that a "dual non-radical mechanism" does not mean the coexistence of two radical species. Rather, it refers to the parallel or sequential participation of two non-radical oxidative routes at the catalytic interface. Typical combinations include $^1O_2$ plus surface-mediated electron transfer, $^1O_2$ plus high-valent metal–oxo species, and electron-transfer-assisted direct oxidative transfer. For example, single-atom Cu–N_4 sites can promote both $^1O_2$ generation and interfacial electron transfer during PMS activation, where the catalyst provides coupled sites for oxidant activation and pollutant oxidation [59]. CuO@biochar has also been reported to induce selective degradation of electron-rich pollutants through the combined contribution of outer-sphere interaction and $^1O_2$-mediated oxidation [56]. In polyvanadate-containing metal–organic frameworks, multi-channel electron-transfer processes can regulate PMS activation and facilitate non-radical pollutant transformation [61]. Nanoconfined Fenton-like catalysts further demonstrate that the dominant non-radical pathway may shift between $^1O_2$ and surface electron transfer depending on confinement environment and active-site exposure [64,65]. In addition, high-valent Fe(IV)=O or Co(IV)=O species may coexist with other interfacial non-radical routes in PMS activation systems, providing another form of coupled non-radical oxidation [58,62,63]. These examples indicate that dual non-radical mechanisms are usually interface-coupled processes governed by active-site configuration, adsorption geometry, electron-transfer capacity, and oxidant activation mode. Therefore, such mechanisms should be identified using multiple lines of evidence, including probe reactions, scavenging tests, EPR, electrochemical analysis, product distribution, and matrix-tolerance experiments.

It should be noted that "low dose" is not an absolute and universal numerical threshold, because catalyst loading and oxidant concentration depend strongly on pollutant concentration, water matrix, reactor configuration, treatment objective, and whether the catalyst is dispersed or immobilized [66,67]. In this review, "low-dose oxidation" refers to an operating regime in which effective contaminant transformation is achieved using lower oxidant input and/or lower available catalytic-site loading than conventional radical-dominated AOPs, while maintaining pathway selectivity, oxidant utilization efficiency, and low byproduct risk [68,69]. Therefore, low dose should be evaluated using several normalized indicators, including catalyst loading, oxidant concentration, oxidant-to-pollutant molar ratio, pollutant removal per unit oxidant, residual oxidant, transformation-product toxicity, and catalyst leaching [70].

As summarized in Table 1, conventional radical-dominated AOPs commonly rely on relatively high oxidant concentrations or strong radical fluxes to ensure rapid pollutant

degradation. In contrast, low-dose mild catalytic systems seek to reduce bulk oxidant consumption by confining oxidative equivalents at the catalyst–pollutant interface or by steering oxidant activation toward $^1O_2$, surface-mediated electron transfer, high-valent metal–oxo species, or direct oxidative transfer [71,72]. Typical dispersed radical AOPs often use transition-metal catalysts at approximately 0.1–1.0 g·L^{-1} and PMS/PDS at approximately 0.5–5 mM, or Fenton/photo-Fenton systems using millimolar to tens-of-millimolar H_2O_2 together with soluble Fe species. By comparison, many mild catalytic studies operate in lower subranges, for example with $\leq$0.1 g·L^{-1} dispersed catalyst, immobilized catalytic interfaces, or $\leq$0.5 mM PMS/H_2O_2 while relying on improved oxidant utilization and interfacial pathway control rather than extensive bulk-phase radical production [73]. These values should be regarded as practical reference windows rather than rigid criteria.

Table 1. Operational interpretation of "low-dose" mild catalysis compared with conventional radical-dominated AOPs.

Catalytic Process Type	Representative Catalyst/Oxidant Window	Dominant Oxidative Behavior	Interpretation in This Review
Homogeneous Fenton/photo-Fenton systems	Fe species commonly in the μM–mM range; H_2O_2 often in the mM to tens-of-mM range	Strong •OH generation; rapid but poorly selective oxidation; usually acidic operation	Conventional radical AOP benchmark; not considered mild solely because Fe dosage is low
Heterogeneous transition-metal/PMS or PDS systems	Solid catalysts commonly around 0.1–1.0 g L^{-1}; PMS/PDS commonly around 0.5–5 mM	SO_4•$^-$/•OH-dominated or mixed radical/non-radical oxidation	Conventional AOP if bulk radical formation dominates; may become mild only if pathway-selective interfacial oxidation is established
Carbon-based non-radical persulfate systems	Catalyst and oxidant dosages can be reduced when electron-transfer or $^1O_2$ pathways dominate	$^1O_2$ generation, surface-mediated electron transfer, or outer-sphere oxidation	Potentially mild when effective removal is achieved with lower oxidant waste and reduced matrix interference
Single-atom or nanoconfined catalytic systems	Often designed to operate with low accessible metal loading, immobilized sites, or reduced oxidant input	High-valent metal–oxo species, $^1O_2$, ETP, or pathway-shifted non-radical oxidation	Mild when active-site regulation improves oxidant utilization, selectivity, and byproduct control
Immobilized catalytic membranes, filters, and hydrogels	Catalyst is described by surface loading, membrane loading, or fixed-bed amount rather than only g L^{-1}; oxidant can be continuously supplied at low concentration	Interfacially confined oxidation under flow-through or continuous operation	Mild when immobilization reduces catalyst release, localizes oxidation, and supports stable low-dose operation
Direct oxidative transfer or interfacial oxidative transfer systems	Oxidant input is minimized relative to conventional radical systems by simultaneous surface activation of pollutant and oxidant	Directional oxidative-equivalent transfer without bulk ROS accumulation	Representative mild catalytic pathway if low-dose operation, reduced byproducts, and interfacial confinement are simultaneously achieved

Importantly, a catalyst that is not "mild" in its intrinsic reaction behavior does not automatically become a mild catalyst simply because it is used at a lower dose. If the

dominant pathway remains bulk-phase •OH/SO_4•$^-$ generation and the process still relies on non-selective radical attack, matrix-insensitive oxidant consumption, or uncontrolled transformation, it should be described as a low-dose conventional AOP rather than a mild catalytic system. Conversely, the same material may exhibit mild catalytic behavior under optimized conditions if its surface state, oxidant dosage, pH, matrix composition, and reactor configuration shift the reaction toward interfacial, selective, low-byproduct pathways. Thus, "low dose" is a necessary but insufficient condition for mild catalysis; mildness should be judged by the combined reaction–material–operation behavior.

From a materials perspective, these non-radical and low-dose oxidation pathways should not be regarded merely as alternative reaction mechanisms, but as material-regulated outcomes of specific active-site structures and interfacial environments. The generation of $^1O_2$ is often associated with electron-rich carbon domains, heteroatom-induced charge redistribution, vacancy-mediated oxygen activation, or dual-site cooperation. Surface-mediated electron transfer requires intimate contact among pollutants, oxidants, and conductive catalytic interfaces, making adsorption configuration and interfacial conductivity critical. High-valent metal–oxo species are more sensitive to the metal coordination environment, ligand field strength, and second-shell modulation. Therefore, the central materials question is how to design active sites and architectures that can selectively stabilize desired reaction intermediates while suppressing uncontrolled radical leakage. This mechanistic understanding provides the basis for the catalyst design strategies discussed in the following section.

3. Design Principles and Activity Control of Mild Catalytic Materials

3.1. Material Platforms and Active-Site Engineering

An ideal catalyst for mild catalysis should meet several key requirements: it should efficiently activate oxidants under near-neutral and mild conditions, preferentially induce the desired reactive oxygen species or interfacial electron-transfer processes, and exhibit good structural stability, recoverability, and long-term operational potential [74–76]. Therefore, the design of mild catalysts should not focus solely on maximizing instantaneous reaction rates. Instead, greater attention should be paid to the synergy among active-site configuration, electronic structure, interfacial mass transfer, and material stability [64]. In the context of materials design, these catalysts can be understood through several key parameters, including metal coordination geometry, heteroatom configuration, vacancy concentration, surface polarity, pore architecture, electrical conductivity, and immobilization stability.

In recent years, the development of single-atom catalysts, heteroatom-doped carbon materials, defect-engineered metal oxides, and immobilized composite catalytic systems has provided important platforms for regulating oxidant activation pathways [77–79].

3.1.1. Single-Atom and Nanocluster Catalysts

The first category is single-atom and nanocluster catalysts. Transition metals such as Fe, Co, and Mn can be anchored as isolated atoms or small clusters on nitrogen-doped carbon, metal–organic-framework-derived carbon, or porous supports. This strategy can substantially improve metal utilization efficiency while enabling precise regulation of PMS, H_2O_2, or O_3 activation through well-defined coordination environments. As shown in Figure 3a [80], Chen et al. [75] constructed Fe–N_4–C_6O_2 single-atom sites through secondary-shell oxygen doping, tuning the local electronic environment without disrupting the primary Fe–N_4 coordination symmetry and thereby enhancing PMS activation and Fe(IV)=O reactivity. This result indicates that the activity of single-atom catalysts is not determined solely by the central metal atom; the secondary coordination sphere, local electric field, metal–ligand bond stability, and local charge distribution can also profoundly

influence reactive-species generation, pathway selectivity, and catalyst lifetime during mild catalysis.

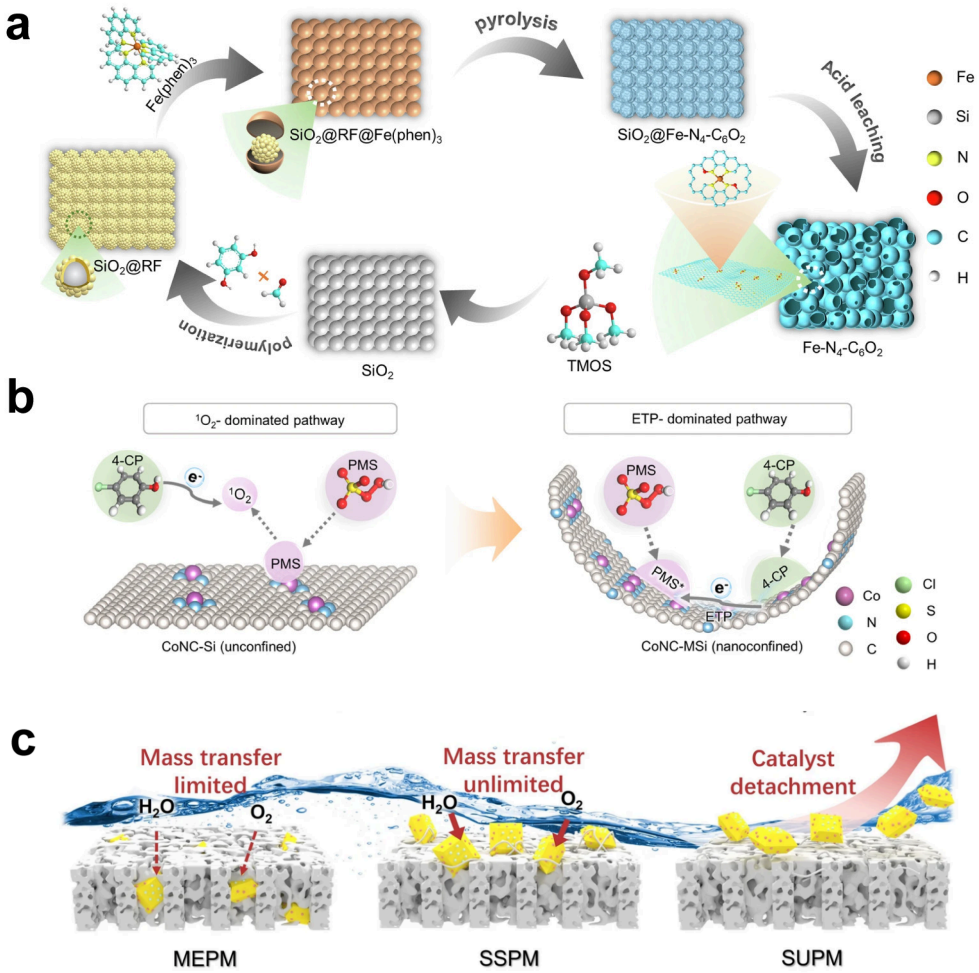


Figure 3. (**a**) Construction of Fe–N_4–C_6O_2 single-atom sites through secondary-shell oxygen coordination for PMS activation and enhanced Fe(IV)=O reactivity. Reprinted by permission from ref. [80]. Copyright (2025) Springer Nature. (**b**) Schematics of the different catalytic pathways for the unconfined and nanoconfined Co SACs. Reprinted by permission from ref. [65]. Copyright (2024) Springer Nature. (**c**) Schematic diagram of the membrane under flow-by operation illustrating the different reactant transfer conditions and catalyst stability. Reprinted by permission from ref. [81]. Copyright (2025) Springer Nature.

More examples further demonstrate the diversity of single-atom, paired-atom, and nanoconfined active sites for mild catalytic oxidation. Single-atom Cu–N_4 sites have been shown to promote both singlet oxygen generation and surface-mediated electron transfer during PMS activation, indicating that isolated metal–N_4 coordination can couple oxidant activation with pollutant electron transfer [59]. Nanoconfined iron single-atom catalytic membranes can enhance PMS activation through a non-radical-dominated pathway, showing how isolated Fe sites and membrane confinement can be integrated to improve oxidant utilization and reduce catalyst release [76,77]. In addition, pair-resolved Fe–M dual-atom catalysts provide another useful platform for programmed PMS activation, because the neighboring metal atoms can tune adsorption geometry, electron distribution, and pathway selectivity [77]. These examples indicate that single-atom and nanocluster catalysts should not be evaluated only by atom utilization efficiency; their coordination environment, neighboring atoms, confinement structure, and immobilized architecture are equally important for determining whether the system behaves as a mild catalytic process.

3.1.2. Heteroatom-Doped Carbon Materials

Carbon-based catalysts and heteroatom-doped systems represent another important class of mild catalytic materials [82,83]. Graphene, carbon nanotubes, biochar, and porous carbon materials possess abundant π-electron structures and tunable surface functional groups, and have therefore been widely used for persulfate activation [84]. For carbon-based catalysts, catalytic function is governed not only by surface area, but also by the electronic structure of the carbon framework; the distribution of graphitic, pyridinic and pyrrolic nitrogen species; the density of edge defects; and the degree of charge polarization induced by heteroatom doping [85]. Introducing heteroatoms such as N, B, S, P, and F, or constructing defects, edge sites, and charge-heterogeneous regions, can alter the electron-donating and electron-accepting properties of carbon materials and thereby influence the competition between radical and non-radical pathways. For example, different nitrogen species may promote PMS adsorption, surface electron transfer, or $^1O_2$ generation, whereas dual-heteroatom sites can further induce local charge polarization and interfacial reaction selectivity. Thus, carbon-based materials should be discussed not as passive supports, but as electronically tunable catalytic interfaces for regulating oxidant activation.

It should also be emphasized that heteroatom-doped carbon materials are not intrinsically mild or non-radical catalysts [86,87]. Depending on the dopant type, defect density, oxygen-containing functional groups, residual metal species, oxidant dosage, and solution conditions, these materials can also behave as conventional ROS-generating catalysts [88]. For example, highly defective carbon surfaces, metal-contaminated carbon materials, or carbon frameworks with abundant redox-active oxygen groups may promote $SO_4^{\bullet-}$/•OH formation and bulk-phase radical oxidation [89]. In contrast, carbon materials are more likely to exhibit mild catalytic behavior when their electronic structure favors outer-sphere PMS activation, surface-mediated electron transfer, or $^1O_2$ generation, while suppressing extensive radical leakage into the bulk solution [90]. Therefore, the key distinction is not whether the catalyst is "carbon-based", but whether heteroatom configuration and surface electronic structure steer oxidant activation toward interfacially confined and pathway-selective oxidation.

Representative examples of heteroatom-doped carbon materials also support this structure-dependent interpretation. N-doped single-walled carbon nanotubes can trigger non-radical catalytic oxidation of phenol, highlighting the role of nitrogen-induced charge redistribution in persulfate activation [32]. Surface nitrogen-modified carbon nanotubes have also been used for catalytic oxidation of organic pollutants, showing that nitrogen functionalities and carbon electronic structure can regulate the generation and utilization of reactive species [84]. More broadly, the roles of structural defects, oxygen-containing groups, and heteroatom dopants in carbon-based catalysts have been systematically shown to determine whether water contaminants are oxidized through radical or non-radical pathways [78]. These examples suggest that heteroatom-doped carbon materials can provide mild catalytic interfaces only when the dopant configuration, defect density, and surface functional groups favor $^1O_2$ generation, outer-sphere PMS activation, or surface electron transfer rather than uncontrolled radical leakage.

3.1.3. Defect-Engineered Metal Oxides and Composites

Transition-metal oxides and composite materials constitute a third major platform for mild catalysis. MnO_2, FeOOH, Co_3O_4, iron–manganese oxides, and amorphous oxides containing oxygen vacancies can activate oxidants through valence cycling and surface defects [72,79,91]. Oxygen vacancies, crystal facets, metal valence distributions, and interfacial heterostructures can influence the adsorption configuration of PMS or H_2O_2, thereby determining whether the system tends to generate free radicals, surface-bound

reactive species, $^1O_2$, or high-valent metal–oxo species [63]. Importantly, defect engineering should not be simplified as increasing the number of vacancies. Excessive or unstable defects may accelerate metal leaching, structural reconstruction, or uncontrolled radical generation. For mild catalysis, the more relevant objective is to construct stable and electronically appropriate defect sites that can adsorb oxidants, guide interfacial charge transfer, and confine reactive intermediates, thereby improving oxidant utilization efficiency and reaction selectivity.

Similarly, defect-engineered metal oxides and composites should not be automatically classified as mild catalysts. Transition-metal oxides with fast redox cycling, excessive oxygen vacancies, unstable surface defects, or high oxidant loading may generate substantial freely diffusing •OH or SO_4•$^-$ and therefore operate as conventional radical-dominated AOP catalysts [92,93]. Mild behavior is more likely when defects or heterointerfaces stabilize adsorbed peroxide intermediates, regulate metal-valence cycling, favor surface-bound reactive species or high-valent metal–oxo species, and reduce radical escape into the bulk phase [28]. The difference is therefore controlled by defect stability, metal redox kinetics, oxidant adsorption geometry, interfacial charge-transfer efficiency, and the balance between surface-confined oxidation and bulk radical release. This distinction is important because simply increasing oxygen-vacancy concentration or metal redox activity may enhance apparent degradation rates, but may also sacrifice selectivity, oxidant efficiency, and catalyst stability.

Beyond the active sites themselves, nanoconfinement has emerged as an important strategy for regulating mild catalytic pathways. As shown in Figure 3b [65], Meng et al. confined Co single atoms within mesoporous structures and found that the reaction pathway shifted from $^1O_2$-dominated oxidation under unconfined conditions to surface electron-transfer-dominated oxidation under nanoconfinement. This process not only increased the local contact probability between pollutants and PMS within the pores, but also altered the surface electronic structure of Co sites, significantly improving PMS utilization efficiency. These findings demonstrate that confined spaces do more than enrich substrates and enhance mass transfer; they can fundamentally reshape oxidant activation pathways, making nanoconfinement a powerful material-design strategy for low-dose and highly selective catalytic transformation.

Taken together, the same material platform can lead to either conventional ROS-dominated AOPs or mild catalytic oxidation, depending on how oxidants are activated and where oxidative equivalents are consumed. In general, the difference is governed by the adsorption mode of PMS or H_2O_2, the lifetime and diffusion distance of reactive species, the extent of radical leakage into the bulk phase, the strength of pollutant–catalyst interaction, the efficiency of interfacial electron transfer, and the stability of active sites under reaction conditions. Therefore, material classification alone is insufficient. A heteroatom-doped carbon, a defect-rich oxide, or a composite catalyst should be considered mild only when pathway diagnostics, matrix-tolerance tests, oxidant-utilization analysis, byproduct evaluation, and stability data collectively indicate selective, interfacially confined, and low-risk oxidation.

Overall, the design of mild catalysts is shifting from conventional "high-activity material screening" toward the integrated regulation of active sites, electronic structures, interfacial environments, and mass-transfer processes [64,74–79]. Through single-atom coordination regulation, heteroatom doping, oxygen-vacancy construction, nanoconfinement, and composite-interface engineering, it is possible to directionally tune the type of reactive species generated, the reaction rate, and byproduct-formation pathways [65]. These advances provide a materials foundation for low-dose, low-byproduct, and ecologically

compatible water remediation. The major material platforms and their structure–function relationships are summarized in Table 2.

Table 2. Material design strategies for mild catalytic oxidation in aquatic remediation.

Material Platform	Key Structural Feature	Dominant Regulation Effect	Possible Mild Pathway	Main Application Advantage	Ref.
Single-atom catalysts	Metal–N_x coordination, second-shell modulation	Controls metal–oxo formation and electron transfer	$^1O_2$, ETP, M(IV)=O	High atom utilization and tunable active sites	[59,63,75,80]
Heteroatom-doped carbon	N/B/S/P/F doping, charge polarization	Regulates PMS adsorption and non-radical pathways	$^1O_2$, ETP	Metal-free or low-metal catalysis	[32,78,84,90]
Defect-engineered oxides	Oxygen vacancies, mixed valence, crystal facets	Stabilizes oxidant adsorption and reactive intermediates	$^1O_2$, M(IV)=O, surface-bound ROS	Robust redox cycling	[63,79,92,93]
Nanoconfined catalysts	Mesopores, core–shell structures, membrane pores	Enhances local concentration and suppresses radical leakage	ETP, non-radical oxidation	Improved oxidant utilization	[16,31,64,65]
Hydrogel catalysts	3D polymer network, water permeability	Reduces catalyst leakage and enriches oxidants	Interfacial oxidation	Recoverable and continuous operation	[94–96]
Floating/immobilized composites	Macroscopic support, buoyant architecture	Enables open-water deployment and recovery	Photocatalytic mild oxidation	In situ applicability	[81,97–99]

3.2. Coupling Material Properties with Reaction Conditions

In addition to the catalyst itself, the activity of mild catalytic systems is also governed by operational parameters such as pH, temperature, oxidant dosage, dosing mode, and reactor configuration [100–102]. First, non-radical pathways generally exhibit broader pH adaptability. For example, $^1O_2$ and surface electron-transfer-mediated processes are typically less dependent on solution pH than •OH generation in conventional Fenton systems, making them more suitable for operation in neutral or weakly alkaline real water matrices [103,104]. Further optimization of pollutant and oxidant adsorption at catalytic interfaces can be achieved by introducing basic sites, modulating surface charge, or constructing hydrophilic/hydrophobic microenvironments on the catalyst surface. The influence of operating conditions is closely related to the intrinsic properties of catalytic materials. For example, surface charge determines pollutant and oxidant adsorption under different pH conditions; pore structure and hydrophilicity affect the diffusion of PMS, H_2O_2, and organic molecules; and electrical conductivity influences interfacial electron transfer. Therefore, pH tolerance, oxidant efficiency, and reaction stability should be interpreted as material-dependent properties rather than only process parameters.

Second, oxidant dosage and dosing strategy are key factors affecting the sustainability of mild catalysis [105,106]. Strictly speaking, oxidant dosage and dosing strategy do not make a catalyst intrinsically mild; rather, they determine whether a catalytic reaction is operated in a mild mode. A catalyst with strong radical-generating capacity may still behave as a conventional AOP catalyst if a high instantaneous oxidant concentration promotes bulk •OH/SO_4•$^-$ production and non-selective oxidation. Conversely, an appropriately designed catalytic material can exhibit mild catalytic behavior only when the oxidant supply is matched with its interfacial activation mode, so that oxidative equivalents are generated

and consumed near the catalyst–pollutant interface rather than being released extensively into the bulk solution. Therefore, oxidant dosing should be regarded as an operational lever that cooperates with active-site design, surface adsorption, mass transfer, and reactor configuration to regulate the reaction pathway. In this sense, "mildness" is a property of the whole catalytic process, rather than a property determined by oxidant dosage alone. Unlike conventional high-intensity oxidation, mild catalytic operation emphasizes effective pollutant transformation at relatively low oxidant dosages. Accordingly, continuous low-dose dosing, slow-release oxidant particles, immobilized oxidant precursors, and in situ H_2O_2 generation are all promising strategies [107,108]. These approaches can avoid excessive instantaneous oxidant concentrations, which often lead to ineffective oxidant consumption, bulk radical leakage, and byproduct risks, while providing more stable reaction conditions for non-radical pathways or interfacial transformation processes.

Third, reactor configuration and catalyst immobilization determine whether mild catalytic systems can move from laboratory-scale powder reactions toward continuous operation and practical engineering applications [109]. As shown in Figure 3c [81], different membrane immobilization strategies can substantially affect reactant mass transfer and catalyst stability. Conventional embedded membrane structures may bury active particles within membrane pores, limiting the transport of H_2O, O_2, or pollutants to active sites. Surface-unfixed catalysts, although highly exposed, are prone to detachment under hydraulic shear. By contrast, surface-self-fixed or binder-free immobilized membranes can maintain active-site exposure while reducing catalyst loss, making them more suitable for continuous-flow treatment and in situ deployment. This design logic indicates that the engineering performance of mild catalytic systems is determined not only by the intrinsic activity of the catalyst, but also by the compatibility among the active material, support, membrane architecture, and hydraulic conditions.

In addition, mild catalysis can be coupled with biological filters, constructed wetlands, membrane separation, coagulation, or ozone contact units [110,111]. For example, mild catalysis may be used as a pretreatment step before biological treatment, where non-radical pathways partially transform recalcitrant pollutants into more biodegradable or less toxic intermediates, followed by further removal in biological units [112,113]. This two-stage "catalytic pretreatment–biological polishing" strategy helps avoid the high chemical and energy demands associated with complete mineralization and is more consistent with the goals of sustainable water remediation. Therefore, the optimization of mild catalytic systems should consider both material structure and operational process [114]. On the one hand, active-site design, electronic-structure regulation, and nanoconfinement can improve reaction selectivity and oxidant utilization efficiency. On the other hand, low-dose dosing, immobilized membrane configurations, and reactor integration can enhance long-term stability and engineering feasibility. Only by integrating material design with system-level optimization can mild catalysis truly serve low-disturbance, low-byproduct, and sustainable aquatic ecosystem restoration.

4. Form-Factor Design, Immobilization, and In Situ Deployment

4.1. Catalyst Immobilization, Support Engineering, and Form-Factor Design

For practical aquatic remediation, catalytic performance is determined not only by nanoscale active sites, but also by the macroscopic form of the material. Powder catalysts often suffer from aggregation, difficult recovery, secondary release, and poor compatibility with continuous-flow or open-water systems. Therefore, immobilization and support engineering should be regarded as form-factor design strategies that translate nanoscale catalytic activity into usable material architectures. Immobilizing catalysts on appropriate supports is a key prerequisite for the in situ application of mild catalysis [94,102,115].

On the one hand, immobilization helps prevent the direct release of nanocatalysts into the environment, thereby reducing potential secondary pollution risks and facilitating catalyst recovery and reuse. On the other hand, the support can endow the catalyst with additional structural functions, such as improving its stability in flowing water, enhancing mass transfer between pollutants and oxidants, or improving its adaptability to sediment–water interfaces, open-water environments, and continuous-flow devices [116]. Therefore, immobilization is not merely a material-shaping strategy, but a critical bridge connecting catalytic activity, process stability, and engineering feasibility.

Common immobilization formats include particle or bead dosing; hydrogel/polymer confinement; and membrane, fiber, or floating units [97]. From a materials-design perspective, these formats correspond to three functional architectures: recoverable packed materials, diffusion-permeable catalytic networks, and spatially deployable catalytic units. As summarized in Figure 4a–c, form-factor engineering can be understood as the coupled design of topological geometry, physical robustness, permeability, and chemical functionality. This design logic helps translate nanoscale catalytic activity into scalable, mechanically stable, flow-through, and deployable purification architectures, thereby addressing the common limitations of powder catalysts, including aggregation, difficult recovery, catalyst detachment, poor hydraulic compatibility, and limited long-term stability.

First, particle- or bead-based catalysts represent recoverable packed materials. These strategies can transform powdered catalysts into millimeter-scale fillers, beads, or fixed-bed materials for river dosing, sediment capping, or continuous-flow reactors. Such particle- or bead-based immobilization strategies are operationally simple, facilitate recovery, are compatible with packed-bed operation, and can reduce the loss of powdered catalysts.

Second, hydrogel-based catalysts function as diffusion-permeable catalytic networks. Hydrogels and polymer networks can confine catalytically active components within three-dimensional polymer frameworks through encapsulation, cross-linking, or coordination interactions. Such networks allow pollutants and oxidants in water to diffuse into internal active regions while spatially restricting catalyst loss and metal leaching. As shown in Figure 5a [95], the PBA/MoS_2@CSH hydrogel exhibited stable long-term continuous-flow performance in a Vis/PMS system. In this system, the chitosan hydrogel network captures PMS and promotes electron transport, while the photoexcited interfacial electric field of the PBA/MoS_2 heterojunction enables stable catalytic oxidation over extended operation. This example suggests that hydrogel immobilization can not only improve material recoverability, but also enhance the sustained performance of mild catalytic systems by locally enriching oxidants, regulating interfacial electron transfer, and suppressing the loss of active components.

Third, membrane-, filter-, fiber-, and floating-unit-based materials represent spatially deployable catalytic architectures. These immobilization structures are more suitable for flow-through treatment, suspended deployment, and open-water in situ remediation. Compared with fixed-bed fillers, membranes and filters can be more readily coupled with water flow and can reduce mass-transfer limitations under continuous operation. As shown in Figure 5b [118], an FePc-modified CNT catalytic filter maintained efficient BPA removal in different natural water matrices and exhibited stable long-term operation with limited Fe leaching. This example demonstrates that immobilized catalytic interfaces can integrate molecularly defined Fe–N_4 active sites, PMS activation, flow-through operation, catalyst retention, and environmental matrix tolerance for mild water remediation. Compared with powdered photocatalysts, floating or suspended materials can reduce sedimentation and the need for post-treatment separation. As shown in Figure 5c [98], the water-hyacinth-inspired self-floating photocatalytic purifier achieves stable flotation through a closed-cell structure and immobilizes TiO_2-based photocatalytic components on a porous substrate.

This design enables improved access to light and oxygen near the water surface while reducing catalyst aggregation, sedimentation, and recovery difficulties associated with powder systems. Such biomimetic floating units provide useful inspiration for in situ photocatalytic remediation in shallow lakes, landscape waters, and slow-flowing rivers.

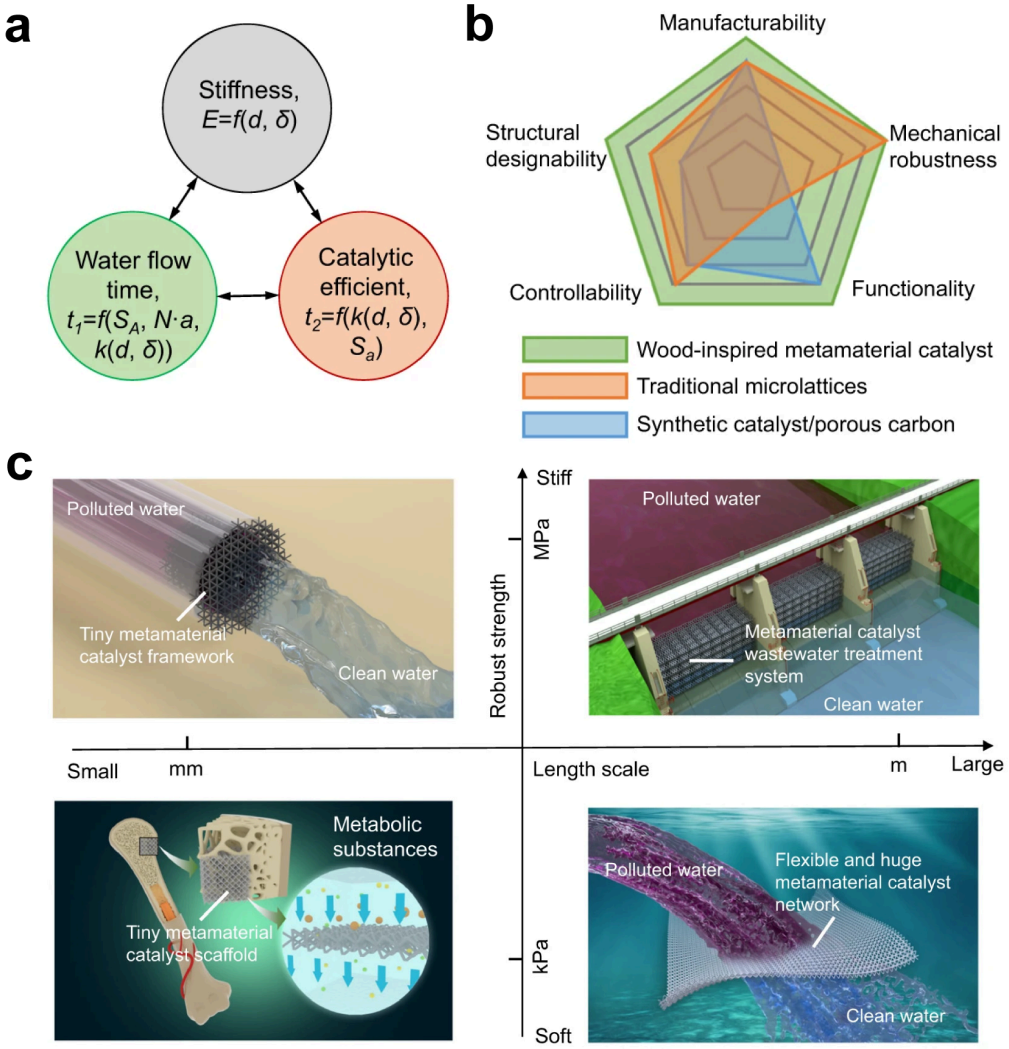


Figure 4. Design logic and application potential of wood-inspired metamaterial catalyst systems for water purification. (**a**) Design paradigm of wood-inspired metamaterial catalyst systems based on the coupled regulation of topological geometry, physical properties, and chemical properties. (**b**) General characteristics comparison of common materials used for water purification, highlighting the importance of structural robustness, permeability, processability, and catalytic functionality. (**c**) Possible applications of wood-inspired metamaterial catalysts across different length scales, illustrating how macroscopic support engineering can enable scalable, flexible, flow-through, and structurally stable catalytic water-purification systems. Reprinted by permission from ref. [117]. Copyright (2024) Springer Nature.

Overall, the choice of immobilization strategy should be tailored to the target application scenario. For continuous treatment or advanced reclaimed-water purification, particle fillers, fixed beds, or hydrogel column devices may be preferred. For open rivers and lakes, floating, suspended, or fiber-supported catalytic units may be more appropriate. For controlling internal pollution from sediments, settleable particles, active capping layers, or bottom-laid composite mats may be more suitable. Through rational design, mild catalysts can maintain localized and controllable activity at remediation sites over extended peri-

ods, enabling the transition from laboratory powder catalysis to in situ, low-disturbance water remediation.

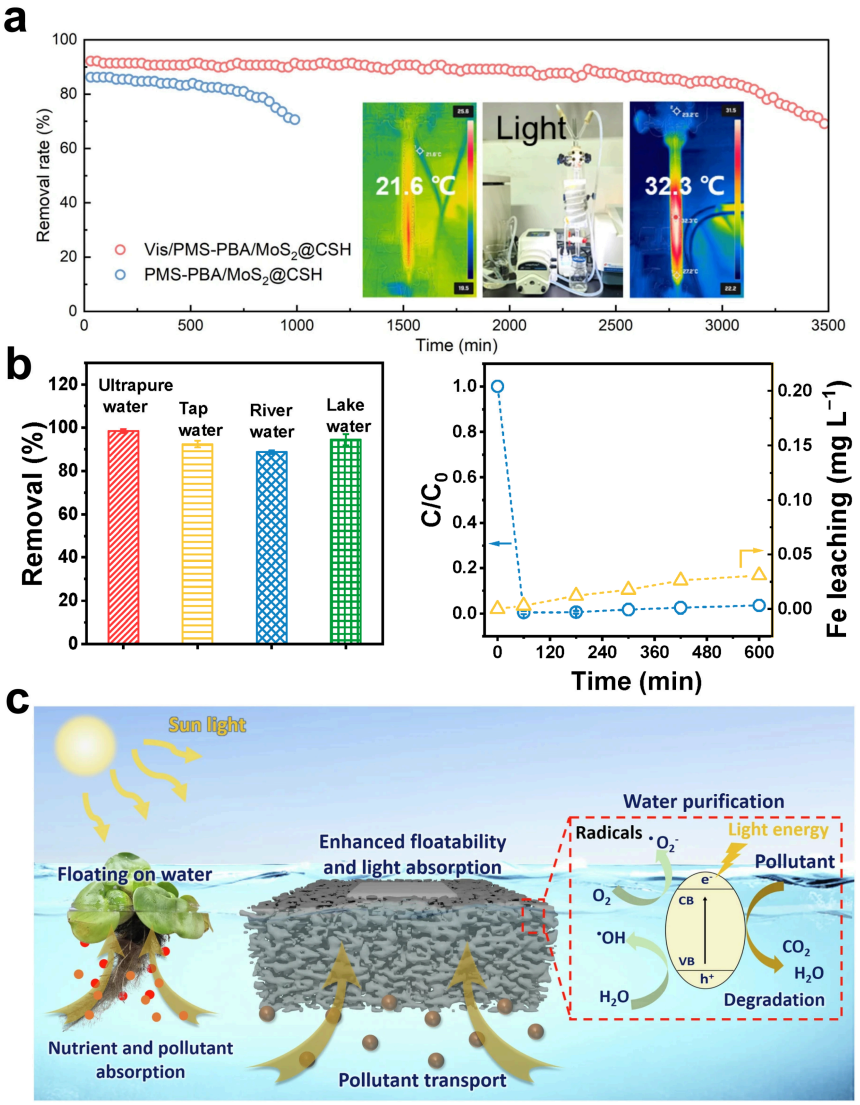


Figure 5. Representative immobilized catalytic architectures for mild water remediation. (**a**) Performance of PBA/MoS_2@CSH in continuous-flow reactions. Reprinted by permission from ref. [95]. Copyright (2023) Springer Nature. (**b**) BPA removal in different natural water matrices under identical single-pass flow-through conditions and long-term operational stability with Fe leaching in the CNT-FePc/PMS catalytic filtration system. Reprinted by permission from ref. [118]. Copyright (2025) MDPI. (**c**) Schematic illustration of the self-floatable photocatalytic water-hyacinth-inspired purifier (WHIP) and its working principle. Reprinted by permission from ref. [98]. Copyright (2025) Springer Nature.

4.2. Representative Scenarios for In Situ Application

Different aquatic remediation scenarios impose distinct requirements on catalytic materials in terms of form factor, mechanical stability, mass transfer, and deployment mode. Therefore, the value of mild catalytic materials should be evaluated not only by intrinsic activity, but also by whether their architecture matches the target aquatic environment. Mild catalysis shows considerable potential in a range of aquatic ecosystem restoration scenarios.

The first scenario is the remediation of black-odorous rivers and lakes [5]. Black-odorous waters are typically associated with anaerobic decomposition of organic matter, sulfide generation, ammonia accumulation, and dissolved oxygen depletion. Direct appli-

cation of high-dose intensified oxidation may improve sensory indicators in the short term, but it may also disturb microbial communities and ecological recovery processes [119,120]. Therefore, a more appropriate strategy is to gradually oxidize reduced malodorous substances and improve the redox status of the water body through low-dose, slow-release, or immobilized catalytic approaches [121]. For example, particulate catalysts or hydrogel beads can be coupled with persulfate, hydrogen peroxide, or percarbonate slow-release systems to establish a relatively mild local oxidative microenvironment. In addition, immobilized catalytic filters provide a complementary flow-through strategy for water treatment and are useful for improving matrix tolerance, operational stability, and catalyst retention under continuous water-treatment conditions.

The second scenario is the control of internal pollution release from sediments. In this review, "internal pollution control" refers to the suppression, immobilization, or transformation of endogenous pollutants released from bottom sediments into the overlying water. In eutrophic lakes, chronically polluted rivers, and black-odorous water bodies, sediments can act as long-term reservoirs of nutrients, reduced substances, and sediment-associated organic contaminants [122,123]. As shown in Figure 6, long-term lake-sediment records across China reveal pronounced historical accumulation trends of total nitrogen (TN) and total phosphorus (TP), indicating that sediments can serve as persistent internal nutrient reservoirs in eutrophic and chronically polluted aquatic systems. Under anoxic, reducing, or hydraulically disturbed conditions, NH^{4+}-N, soluble reactive phosphorus, Fe(II), Mn(II), sulfide, dissolved organic matter, and sediment-bound micropollutants may be released across the sediment–water interface, thereby sustaining eutrophication, oxygen depletion, odor generation, and secondary water-quality deterioration even after external inputs are reduced [124]. Therefore, internal pollution control is not limited to the removal of pollutants already present in the water column; it also aims to regulate interfacial fluxes from sediments and prevent the continuous regeneration of pollutants.

Mild catalytic materials can be deployed on the sediment surface as active capping layers, settleable particles, catalytic mats, or bottom-laid composite units. Compared with dredging or direct high-dose oxidant addition, these in situ strategies are less disruptive and can provide more sustained interfacial control [126,127]. Previous studies have shown that active capping materials, including activated carbon, zeolite, organoclay, apatite, red soil, aluminum salts, lanthanum-modified bentonite, and iron/calcium-based mineral composites, can reduce contaminant mobility or suppress the release of nitrogen and phosphorus from sediments [128,129]. For mild catalytic systems, similar form-factor logic can be used to construct localized oxidative microenvironments at the sediment–water interface. Such systems may oxidize reduced odor-causing substances, regulate sediment redox status, immobilize phosphorus, and transform labile organic contaminants while avoiding the strong ecological disturbance associated with bulk-phase oxidation.

The possible terminal or near-terminal products should also be considered when evaluating internal pollution control [130]. For example, reduced sulfur species such as HS^-/S^{2-} may be oxidized to elemental sulfur or SO_4^{2-}; Fe(II) and Mn(II) can be converted to Fe(III)/Mn(III/IV) oxyhydroxides, which may further bind phosphate; ammonium and nitrogen-containing organics may be transformed through coupled nitrification–denitrification to NO^{3-} and eventually N_2; and organic contaminants may first form low-molecular-weight carboxylic acids, such as formic, acetic, oxalic, malonic, or succinic acids, before further mineralization to CO_2 and H_2O under sufficient oxidation or subsequent biological polishing. Importantly, phosphorus is not "degraded" in the same way as organic pollutants, but is immobilized as more stable Fe/Al/Ca/La-bound phosphate phases. Therefore, mild catalytic control of internal pollution should be evaluated by both pollutant transformation and interfacial flux suppression, including changes in NH^{4+}-N,

soluble reactive phosphorus, sulfide, dissolved organic carbon, redox potential, catalyst leaching, and benthic ecological responses.

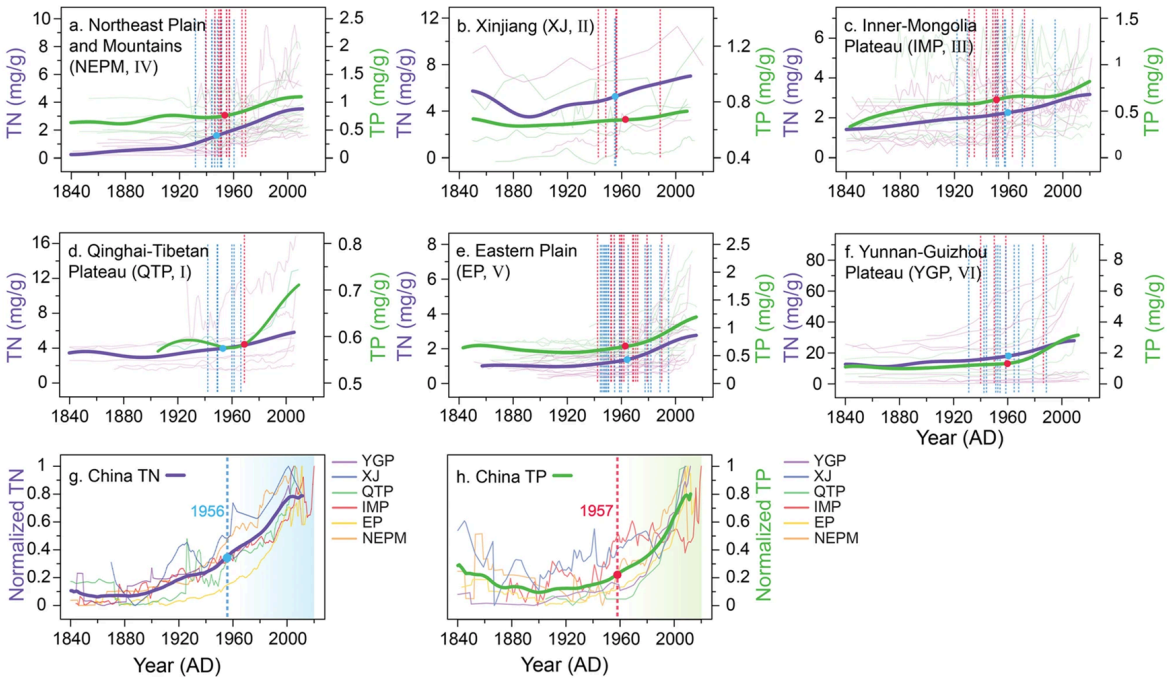


Figure 6. Historical accumulation trends and structural transition nodes of total nitrogen (TN) and total phosphorus (TP) in lake sediments across China. (**a**–**f**), Time-series records of TN and TP in lake sediments from six lake districts in China. Thin dashed curves represent individual lake records, whereas bold curves represent district-averaged trends after 20-year smoothing. Purple and green curves indicate TN and TP, respectively. Vertical dashed lines indicate transition nodes for individual lakes, with blue and red denoting TN and TP transition nodes, respectively. (**g**,**h**), District-averaged and national-scale smoothed trends of TN and TP in Chinese lake sediments. Blue and red dots indicate transition nodes of the corresponding averaged curves. This figure highlights the long-term accumulation of sedimentary N and P and provides evidence for considering sediments as important internal nutrient reservoirs in eutrophic and chronically polluted aquatic systems. Reprinted by permission from ref. [125]. Copyright (2024) Springer Nature.

The third scenario is the removal of trace organic contaminants and emerging pollutants. Drinking-water sources, reclaimed-water-fed rivers, and urban landscape waters often contain low concentrations of antibiotics, pesticides, endocrine-disrupting compounds, dyes, and other micropollutants [131,132]. In such settings, the goal of mild catalysis should not be complete mineralization alone; instead, emphasis should be placed on selective transformation, toxicity reduction, and ecological risk control under low-dose conditions. Hydrogel-immobilized catalysts can be used for continuous-flow advanced treatment, whereas floating photocatalytic units can be applied for open-water in situ purification [133,134]. These two formats represent device-based continuous treatment and open-environment in situ remediation, respectively.

4.3. Challenges in Immobilized and In Situ Applications

Although catalyst immobilization facilitates recovery and long-term operation, its practical application still faces challenges related to mass transfer, stability, and environmental adaptability [94,99,117]. On the one hand, pollutants and oxidants must diffuse into support pores, hydrogel networks, or membrane-surface active regions before reaction can occur, which may impose mass-transfer limitations on reaction rates. In particular, for hydrogel-embedded or particle-packed systems, poorly designed pore structures, hydrophilicity, or

surface charge may lead to active-site shielding, insufficient oxidant utilization, or excessive local concentration gradients. The continuous-flow hydrogel system shown in Figure 5a indicates that a rational three-dimensional polymer network can promote PMS capture and electron transport, but this advantage depends on the matching among hydrogel structure, irradiation conditions, flow rate, and pollutant diffusion behavior.

On the other hand, immobilized structures must remain stable under long-term hydraulic shear, sediment deposition, biofilm growth, and complex water chemistry [96,135]. For fixed-bed or bead-based systems, filler compaction, pore blockage, and leaching of active components must be prevented. For floating photocatalytic units, stable flotation under both static and dynamic hydraulic conditions must be ensured, while detachment of photocatalytic materials and surface coverage by algae or biofilms should be avoided. The WHIP system shown in Figure 5c enhances buoyancy and water-surface stability through a biomimetic closed-cell structure, providing a useful strategy for addressing the sedimentation and recovery difficulties of powdered photocatalysts. Nevertheless, its resistance to fouling, aging, and mechanical degradation during long-term field operation remains to be further evaluated.

In addition, in situ remediation must also consider oxidant storage and dosing safety, seasonal temperature variation, light fluctuation, and interference from natural organic matter and inorganic ions in real water matrices. Overall, although in situ mild catalysis is highly promising, laboratory-scale powder-catalysis results cannot be directly extrapolated to river and lake ecosystems. Future research should systematically address support durability, catalyst detachment risk, controlled oxidant release, deployment configuration, and compatibility with ecological engineering measures, thereby enabling long-term, safe, and sustainable aquatic ecosystem restoration.

5. Environmental Safety and Ecological Compatibility of Mild Catalytic Materials

5.1. Impacts on Microbial Communities

The ultimate goal of aquatic ecosystem restoration is to recover healthy ecological functions; therefore, assessing the impacts of mild catalysis on indigenous microbial communities is essential [136,137]. Microorganisms are not only passive biological receptors, but also key drivers of organic-matter degradation, nitrogen transformation, phosphorus cycling, sulfur oxidation–reduction, biofilm formation, and water self-purification. Conventional strong oxidants may rapidly reduce pollutant concentrations, but they can also damage microbial cell membranes, extracellular polymeric substances, enzymes, DNA, and redox-active metabolic systems, thereby temporarily suppressing microbial abundance, diversity, and ecological function [138,139]. Such disturbances may be particularly important for functional microbial groups involved in nitrification, denitrification, organic-carbon mineralization, sulfate reduction, sulfur oxidation, and biofilm-mediated nutrient cycling.

For mild catalytic systems, the expected advantage is not that microorganisms are completely unaffected, but that oxidative pressure is more localized, lower in intensity, and more compatible with ecological recovery. Non-radical or interfacially confined pathways may reduce the exposure of non-target microorganisms to freely diffusing strong radicals, while low-dose oxidant delivery can avoid prolonged oxidative stress in the bulk water phase. However, this assumption must be experimentally verified. Evaluation should include microbial biomass, cell viability, extracellular polymeric substances, membrane integrity, enzyme activity, and community structure. High-throughput 16S rRNA sequencing, metagenomics, or metatranscriptomics can be used to analyze changes in microbial diversity and community composition, while quantitative PCR can be used to track functional genes related to nitrogen cycling, such as amoA, nxr, nirS/nirK, nosZ, narG, and

nrfA, as well as genes associated with carbon degradation, sulfur cycling, oxidative stress response, and antibiotic resistance when relevant [136,137].

Previous studies have shown that in situ chemical oxidation may temporarily suppress the abundance and diversity of indigenous microorganisms, but such effects are not necessarily irreversible [138]. After oxidants are consumed, pollutant loads decrease, and physicochemical conditions recover, some indigenous microbial communities may gradually rebound. Analogous with river and lake restoration, low-dose mild catalysis may initially inhibit sensitive microbial populations, especially nitrifiers, algae-associated bacteria, or biofilm-forming organisms. However, if the process reduces pollutant toxicity, avoids persistent transformation products, limits catalyst leaching, and does not maintain long-term oxidative stress, it may create more favorable conditions for the recovery of self-purifying microbial communities. Therefore, both short-term inhibition and long-term recovery should be considered.

The response of microbial communities may also depend strongly on catalyst form and deployment mode. Dispersed nanocatalysts or insufficiently immobilized particles may interact directly with microbial cells and biofilms, causing membrane damage, metal-ion stress, oxidative stress, or changes in extracellular polymeric substances. In contrast, immobilized catalysts, catalytic membranes, hydrogels, or floating units may reduce direct microbial exposure by confining active sites within macroscopic supports. Nevertheless, immobilized materials can still affect microbial colonization, biofilm growth, and local redox microenvironments. Some catalytic surfaces may suppress biofouling, whereas others may serve as attachment substrates for biofilm development. Therefore, catalyst leaching, particle release, surface aging, and biofilm formation should be monitored together with microbial community responses.

The freshwater microcosm assessment framework shown in Figure 7a provides an important reference for mild catalysis research. Such a framework not only considers reductions in micropollutant concentrations, but also evaluates whether ecological communities are reorganized and whether biological functions recover after treatment [41]. For future studies, microbial assessment should include at least three levels: (i) structural indicators, such as alpha diversity, beta diversity, and relative abundance of dominant taxa; (ii) functional indicators, such as nitrification and denitrification genes, organic-matter degradation capacity, respiration activity, and enzyme activity; and (iii) ecological recovery indicators, such as the restoration of self-purification capacity, algae–bacteria balance, and benthic microbial function. In this way, mild catalysis can be evaluated not only by contaminant removal, but also by whether it reduces ecological pressure while preserving or restoring key microbial functions.

5.2. Algae–Bacteria Balance and Ecological Function

In eutrophic waters, algae and bacteria jointly constitute the foundation of primary production and biogeochemical cycling, and their interactions directly influence water-quality stability and ecosystem health [136,137,141]. Excessively strong chemical oxidation may reduce algal cell abundance or improve water transparency in the short term. However, if extensive algal cell rupture occurs, intracellular organic matter, nitrogen and phosphorus nutrients, and algal toxins may be released, potentially causing secondary pollution or bloom recurrence [134,141,142]. Therefore, in eutrophic or algae-impacted waters, mild catalysis should not simply aim for complete algal eradication. Instead, it should emphasize low-dose operation, selective transformation, and regulation of ecological processes.

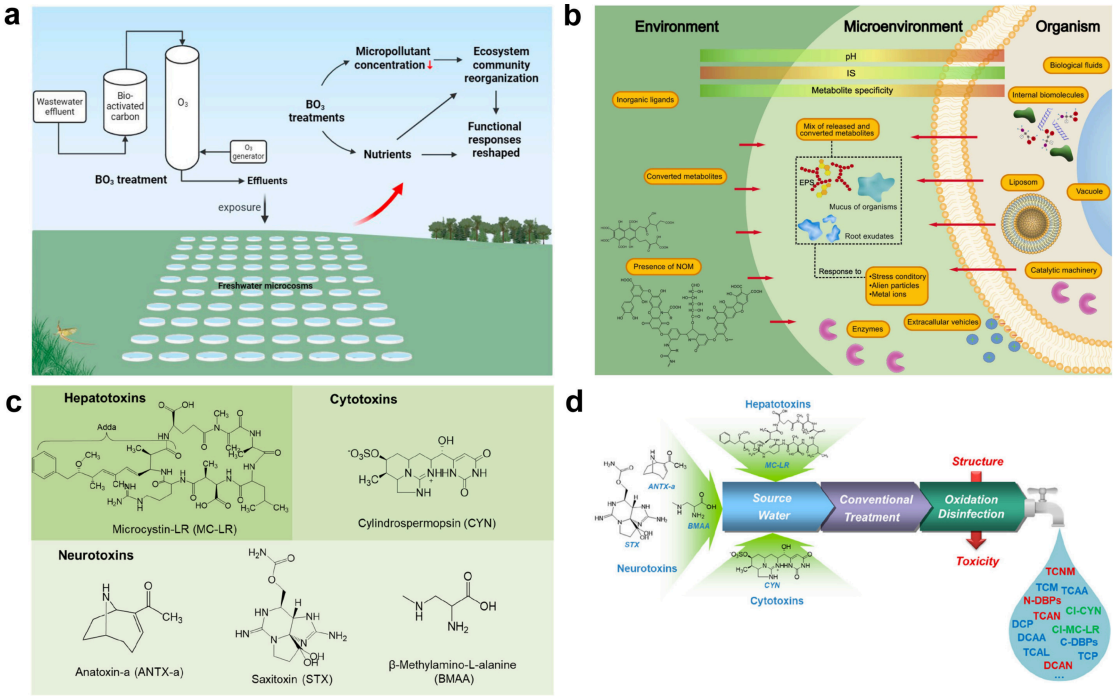


Figure 7. (**a**) Freshwater microcosm-based ecological assessment. Reprinted by permission from ref. [41]. Copyright (2026) Elsevier. (**b**) The variability of conditions between the environment and biota, which may determine the transformation of ENPs. Reprinted by permission from ref. [140]. Copyright (2025) Elsevier. (**c**) Examples of algal toxins that are hepatotoxic, cytotoxic, or neurotoxic. (**d**) Transformation of Algal Toxins during the Oxidation/Disinfection Processes of Drinking Water. Reprinted by permission from ref. [141]. Copyright (2023) American Chemical Society.

Several catalyst examples illustrate how catalytic materials have been used for algal control, cyanotoxin degradation, or microbial regulation. TiO_2 photocatalysis is one of the most representative systems for cyanobacteria-impacted waters. For example, solar-driven TiO_2 photocatalysis has been used to destroy Microcystis aeruginosa cells and simultaneously degrade intracellular and extracellular microcystin-LR and cylindrospermopsin [142]. Immobilized TiO_2 on glass tubes have also been applied in a continuous-flow UV-A photocatalytic reactor for the degradation of microcystin-LR and cylindrospermopsin, providing a relevant example of catalyst immobilization for cyanotoxin treatment [143]. In addition, a melamine-sponge-supported Bi_2O_3@Cu-MOF floating photocatalyst was reported for the inactivation of Microcystis aeruginosa, showing the potential of floating catalytic materials for open-water algal control [134]. Beyond algal systems, silver single atoms and nanoparticles on floatable monolithic photocatalysts, MoO_3^-x-driven Cu^+-doped nanozymes, and aromatic-ketone-derived photosensitizers have been used for microbial disinfection or antibacterial regulation [139,144,145]. These examples indicate that catalytic materials can regulate algal or bacterial cells through interfacial ROS generation, photocatalytic oxidation, or photodynamic pathways. However, for aquatic ecosystem restoration, such antimicrobial or algicidal activity should be carefully balanced against the preservation of beneficial microbial communities and self-purification functions.

Typical algal toxins include hepatotoxic, cytotoxic, and neurotoxic compounds. Figure 7c summarizes representative algal toxins, including microcystin-LR (MC-LR), cylindrospermopsin (CYN), anatoxin-a (ANTX-a), saxitoxin (STX), and β-N-methylamino-L-alanine (BMAA) [141]. These compounds have complex structures and distinct toxicological targets, indicating that safety evaluation during oxidative treatment should not rely solely on the decrease in parent-toxin concentrations. Mild catalysis can reduce algal toxins and taste-and-odor compounds through low-dose oxidation while minimizing large-

scale algal cell lysis, thereby limiting the instantaneous release of intracellular toxins and nutrients [141–143].

At the same time, oxidation processes may alter both the structures and toxicities of algal toxins. As shown in Figure 7d [141], during conventional oxidation/disinfection, algal toxins can undergo structural transformation, which may be accompanied by the formation of transformation products or disinfection byproducts. Therefore, structural destruction does not necessarily equate to complete detoxification. For mild catalysis, the ideal objective is to selectively oxidize key toxic functional groups in a controllable manner while reducing the formation of halogenated byproducts and highly toxic intermediates. If transformation products with ecological risks are still detected after treatment, further polishing processes, such as biological activated carbon, constructed wetlands, or biofiltration, should be considered [110,111,113].

In addition, mild catalysis may facilitate downstream biological processes by improving water biodegradability. For example, some non-radical oxidation pathways can transform macromolecular recalcitrant organics into small organic acids or other intermediates that are more readily utilized by microorganisms, thereby enhancing subsequent microbial degradation [110–113]. This "chemical pre-transformation–biological polishing" strategy may help achieve a more reasonable balance between pollutant abatement and ecological function recovery.

5.3. Ecotoxicity and Safety Assessment

Among all ecological concerns, catalyst-derived risks are particularly relevant to materials research [140,146]. Mild catalytic remediation must ensure that no new ecological risks are introduced through catalyst leaching, particle detachment, surface transformation, or persistent transformation products [146,147]. Therefore, systematic toxicity monitoring during and after treatment is essential. First, the toxicity of oxidative intermediates should be carefully considered [130,148]. Although mild catalysis is expected to reduce byproduct risks associated with non-selective attack by strong radicals, different pollutants may still generate complex transformation products under different reactive species. In some cases, a decrease in parent pollutant concentration does not necessarily indicate a reduction in overall toxicity; certain intermediates may exert stronger acute or chronic effects on algae, daphnids, fish embryos, or microorganisms. Therefore, evaluation of mild catalytic systems should include non-target mass spectrometric screening, transformation-pathway analysis, and multi-trophic-level toxicity assays, rather than relying solely on removal efficiency or mineralization [149].

Second, catalyst leakage and environmental transformation risks should be addressed. During long-term operation, immobilized catalysts may undergo particle detachment, metal leaching, surface redox transformation, or complexation with natural organic matter [146,147]. Figure 7b illustrates the environmental and biological microenvironmental changes that engineered nanoparticles may experience when moving from environmental media into biological microenvironments or organisms. These changes include variations in pH, ionic strength, metabolites, natural organic matter, extracellular polymeric substances, enzymes, and biomolecules, all of which can influence nanoparticle transformation, migration, exposure routes, and potential toxicity. Surface coating, polymer modification, and matrix-induced aging may further alter particle stability, aggregation behavior, and biointerface interactions [150]. For mild catalytic materials, even if high activity and stability are observed in short-term experiments, long-term leaching, particle aging, and biointerface interactions must still be assessed in real water matrices [151,152].

Third, an integrated ecological-impact assessment framework should be established. A comprehensive evaluation should include target pollutants and transformation products,

acute and chronic ecotoxicity, catalyst and metal-component release, nanoparticle detachment, surface oxidation-state evolution, structural aging, microbial community structure, nitrification and denitrification functional genes, algal growth status, benthic organism responses, and basic physicochemical water-quality parameters [153,154]. Compared with conventional "one-shot" strong oxidation strategies, mild catalysis is more consistent with the concept of low-disturbance ecological engineering. Its advantages should be reflected in pollutant-risk reduction, no increase in ecotoxicity, preservation of key functional microbial groups, and recovery of water self-purification capacity. In other words, the ecological compatibility of mild catalysis should not be presumed, but should be verified through the multi-level assessment framework illustrated in Figure 7a–d.

6. Future Perspectives and Challenges

As a more controlled and materials-oriented extension of advanced oxidation, mild catalysis holds substantial promise for aquatic ecosystem restoration. Looking ahead, its future development and major challenges can be summarized as follows.

6.1. Precision Regulation

A central scientific challenge for mild catalysis is how to precisely generate the desired reactive species in complex aquatic environments while avoiding unnecessary side reactions [155,156]. Different remediation stages may require different oxidative intensities. For example, a certain radical contribution may be beneficial at the initial stage to rapidly decrease high contaminant loads, whereas non-radical pathways may be more desirable at later stages to maintain selective treatment. This calls for the development of intelligent regulation strategies, such as real-time modulation of the radical/non-radical balance by tuning catalyst surface electric fields or applying small external currents [157,158]. Another promising direction is the design of self-adaptive catalysts that respond to environmental conditions. In addition, more reliable methods are needed to distinguish radical and non-radical contributions, together with online monitoring tools that can dynamically evaluate and optimize mild catalytic processes in practical systems.

6.2. Ecological Synergy and Multilevel Integration

Mild catalysis should be integrated with other components of ecological restoration to form coupled engineering–ecological remediation systems [159,160]. Future studies may increasingly explore strategies such as coupling mild catalytic units with constructed wetlands or biofilters, where chemical oxidation first alleviates the toxicity barrier of pollutants and subsequent ecological processes complete degradation and nutrient cycling [161]. Mild catalysis may also be integrated with aquatic plant remediation. For instance, root exudates may enhance catalyst activity, while trace amounts of H_2O_2 generated during catalytic reactions may stimulate plant stress tolerance, thereby forming a positive feedback loop in which ecological processes promote catalysis and catalysis supports ecological recovery. At the watershed scale, mild catalysis should be incorporated into integrated water-environment management strategies together with source control, sewage interception, ecological buffer zones, and habitat restoration [162]. Such integration can support multiple objectives, including organic contaminant removal, eutrophication mitigation, and habitat recovery. Regional water governance should therefore develop catalyst–ecology combined remediation schemes tailored to specific watershed characteristics to improve overall treatment efficiency and sustainability.

6.3. Engineering Scale-Up, Regeneration, and Techno-Economic Considerations

Many mild catalytic systems remain at the laboratory or bench scale, and their transition to practical engineering applications will encounter new challenges [163,164]. These

include large-scale catalyst synthesis and stable immobilization, the influence of complex field water chemistry on catalytic performance, regeneration after fouling or poisoning, and the reliability of long-term operation. At present, most mild catalytic materials, especially single-atom catalysts, nanoconfined catalysts, catalytic hydrogels, floating photocatalysts, and catalytic reactive membranes, are still mainly at the laboratory, pilot, or prototype level [165,166]. Although some related water-treatment units, such as ozonation, biological activated carbon, constructed wetlands, and membrane-based advanced treatment, have been implemented at larger scales, the full-scale application of mild catalytic materials for in situ aquatic ecosystem restoration remains limited. Therefore, more pilot-scale and demonstration studies are needed to identify scale-up rules, operating windows, failure modes, and practical bottlenecks [167].

Regeneration is a central issue for the long-term use of mild catalysts. In real water bodies, active sites may be blocked by natural organic matter, suspended solids, carbonate, phosphate, chloride, biofilms, algal residues, or transformation products [168]. Immobilized catalysts may also suffer from pore blockage, surface passivation, particle detachment, metal leaching, and mechanical aging [169,170]. Therefore, regeneration strategies should be designed according to the material form and deactivation mechanism. For membrane or filter catalysts, hydraulic backwashing, air scouring, intermittent flow reversal, low-dose oxidant flushing, or electrochemical self-cleaning may be used to remove foulants and recover active interfaces. For hydrogel, bead, or packed-bed catalysts, mild chemical washing, pH adjustment, light-assisted regeneration, or modular replacement may be more practical. For floating or bottom-laid catalytic units, in situ regeneration should avoid strong chemical cleaning and instead rely on sunlight-driven self-cleaning, aeration, periodic retrieval, or replaceable modules. Importantly, regeneration should not introduce new ecological risks; metal leaching, particle detachment, residual oxidant, and toxicity after regeneration should be evaluated together with catalytic activity recovery.

The cost of mild catalysts should also be compared with that of conventional ROS-generating catalysts. Traditional radical-based systems, such as homogeneous Fenton, heterogeneous transition-metal/PMS systems, and other strong ROS catalysts, often have advantages of simple reagents, low material cost, and rapid pollutant degradation [171]. However, their total operating cost can increase because of high oxidant demand, pH adjustment, sludge generation, radical quenching by water matrices, byproduct control, and post-treatment requirements. In contrast, mild catalysts may require higher initial material costs because of more sophisticated active-site regulation, immobilization, membrane fabrication, or support engineering. Nevertheless, their potential economic advantages lie in lower oxidant input, improved oxidant utilization, reduced matrix scavenging, better catalyst retention, lower byproduct risk, and longer service life. Therefore, the cost comparison should not be based only on catalyst price per gram, but should include normalized pollutant removal per unit oxidant, catalyst lifetime, regeneration frequency, metal leaching control, energy input, post-treatment demand, ecological-risk management, and replacement cost [172].

From a materials perspective, low-cost and scalable catalyst synthesis methods should be developed, such as one-pot synthesis, low-temperature preparation, use of industrial byproducts or biomass-derived carbon supports, and scalable fabrication of catalytic membranes or monolithic units [163,164]. Recent progress in reactive membrane design and nanoconfined catalytic architectures suggests the possibility of improving catalyst utilization, mass-transfer matching, and long-term operational stability while reducing secondary release risks [173,174]. However, before mild catalysts can be considered mature for large-scale water remediation, future studies should report not only degradation efficiency, but also catalyst productivity, material cost, oxidant consumption, regeneration efficiency, life-

time, leaching, energy demand, and performance in real water matrices. Only when mild catalysts remain efficient, regenerable, durable, safe, and economically competitive under realistic operating conditions can they move from laboratory and pilot demonstrations toward practical large-scale application.

6.4. Standards, Regulation, and Environmental Safety

The large-scale deployment of emerging technologies requires appropriate standards and regulatory support [175,176]. Current engineering guidelines for advanced oxidation mainly focus on conventional processes such as Fenton oxidation and ozonation. For emerging systems such as mild catalysis, technical guidelines and safety protocols are urgently needed. These should define appropriate dosing ranges and safety thresholds for different remediation scenarios, as well as harmlessness criteria for treated water, including byproduct concentrations and biological toxicity. Regulatory frameworks should also incorporate micropollutant removal and ecological impacts into water-environment assessment systems [177,178]. A robust monitoring and evaluation framework is necessary to ensure that each step of mild catalytic application is evidence-based and risk-controlled. In addition, public communication and stakeholder engagement should be strengthened to improve understanding of the ecological and environmental benefits of mild catalysis and to facilitate practical implementation.

7. Conclusions

Mild catalytic materials are emerging as an important class of functional materials for low-disturbance aquatic remediation. Unlike conventional high-intensity oxidation systems that primarily depend on strong radical species and high oxidant dosages, mild catalytic systems emphasize controlled oxidant activation, selective pollutant transformation, reduced byproduct formation, and improved compatibility with aquatic ecological processes. From a materials perspective, the key to achieving mild catalysis lies in the rational regulation of active sites, electronic structures, interfacial environments, and mass-transfer pathways. Defect-engineered metal oxides, heteroatom-doped carbon materials, single-atom catalysts, nanoconfined systems, catalytic membranes, hydrogels, and floating or immobilized composites all provide versatile platforms for steering radical and non-radical pathways.

This review highlights that catalyst composition, coordination environment, oxygen vacancies, heteroatom configuration, local electric fields, and confined reaction spaces can profoundly influence the type, amount, and lifetime of reactive species generated during oxidant activation. Non-radical or low-intensity oxidative pathways, including singlet oxygen generation, surface-mediated electron transfer, high-valent metal–oxo species, and direct oxidative transfer processes, offer opportunities to improve oxidant utilization and reaction selectivity while reducing unnecessary oxidative consumption in bulk water. These mechanisms are particularly relevant to the treatment of complex aquatic systems, where natural organic matter, halides, carbonate species, suspended solids, and biological components may strongly interfere with conventional radical oxidation.

In addition to intrinsic catalytic activity, material form and architecture are critical for practical implementation. Immobilized catalysts, catalytic membranes, hydrogel networks, fixed-bed particles, and floating photocatalytic units can improve catalyst recovery, reduce particle release, enhance operational stability, and facilitate in situ or continuous-flow applications. However, these material configurations also introduce new challenges, such as mass-transfer limitations, pore blockage, active-site shielding, metal leaching, biofouling, mechanical aging, and long-term structural instability. Therefore, future material design

should not only pursue high reaction rates, but also balance catalytic efficiency, durability, recyclability, environmental safety, and compatibility with real aquatic conditions.

Looking forward, the development of mild catalytic materials should move toward more precise active-site regulation, controllable oxidant delivery, adaptive reaction interfaces, and integrated catalytic–ecological treatment systems. Standardized evaluation methods are also needed to assess not only pollutant removal, but also transformation-product toxicity, catalyst leakage, microbial community response, algae–bacteria balance, and long-term ecological effects. By linking materials chemistry with water remediation and ecological safety, mild catalytic materials may provide a promising route toward sustainable aquatic ecosystem restoration.

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Abbreviations

The following abbreviations are used in this manuscript:

AOPs	Advanced oxidation processes
PMS	Peroxymonosulfate
PDS	Peroxydisulfate
ROS	Reactive oxygen species
$^1O_2$	Singlet oxygen
•OH	Hydroxyl radical
SO_4•$^-$	Sulfate radical
ETP	Electron-transfer pathway
DOTP	Direct oxidative transfer process
SACs	Single-atom catalysts
Co SACs	Cobalt single-atom catalysts
CNTs	Carbon nanotubes
MOF	Metal–organic framework
TN	Total nitrogen
TP	Total phosphorus
NH_4^+-N	Ammonium nitrogen
NO_3^-	Nitrate
MC-LR	Microcystin-LR
CYN	Cylindrospermopsin
ANTX-a	Anatoxin-a
STX	Saxitoxin
BMAA	β-N-methylamino-L-alanine
ENPs	Engineered nanoparticles
EPR	Electron paramagnetic resonance

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