



Graphene Oxide-based Adsorbents for Pulp and Paper Wastewater Treatment: A Comprehensive Review

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ABSTRACT

The pulp and paper industry generates substantial volumes of complex wastewater containing lignin, dyes, chlorinated organics, and heavy metals, posing significant environmental challenges. Traditional treatment methods are often ineffective in eliminating these persistent pollutants, necessitating advanced solutions. This comprehensive review evaluates the role of graphene oxide (GO) and its derivatives in pulp and paper industry wastewater treatment. Based on a broad synthesis of peer-reviewed studies published between 2015 and 2024, the analysis focuses on pollutant-specific performance, adsorbent types, and treatment efficiencies. The findings demonstrate that GO-based adsorbents outperform conventional materials, achieving removal efficiencies exceeding 90% for lignin, dyes, and heavy metals under optimal pH and contact time conditions. Functionalized GO variants such as GO-ZnO, amine-GO, and GO-polymer hybrids exhibit enhanced adsorption capacities (up to 210 mg/g), faster kinetics, and moderate regeneration potential over 3–5 cycles. Key mechanisms include π - π stacking, hydrogen bonding, and electrostatic attraction. However, limitations remain regarding pilot-scale validation, particularly challenges related to material recovery, process integration, hydraulic performance, and cost scalability as well as long-term stability issues such as adsorbent fouling, structural degradation, and performance loss under continuous operation, along with concerns related to the environmental fate and potential eco-toxicity of GO nanomaterials. The review identifies critical knowledge gaps in mixed pollutant remediation, toxicity assessments, and real effluent application. Future directions recommend green synthesis of GO composites, integration with membrane and photocatalytic systems, and pilot-scale trials under continuous flow. The study underscores GO's potential as a next-generation material for sustainable, scalable, and multi-functional treatment of pulp and paper industrial effluents.

Keywords: Adsorption; Graphene oxide; Industrial effluents; Nanomaterials; Wastewater treatment.

1. INTRODUCTION

The pulp and paper industry is one of the largest and most water-intensive industrial sectors worldwide, producing more than 420 million tonnes of paper and paperboard annually. India alone accounts for approximately 18 million tonnes of this production, ranking as the fifth-largest producer globally (Toczyłowska-Mamińska, 2017; Singh *et al.* 2023). Despite its economic importance, the industry exerts a significant environmental burden, primarily due to excessive water consumption and wastewater generation. During paper manufacturing, approximately 60–100 m³ of wastewater is discharged per tonne of paper produced, depending on the process and level of recycling. These effluents contain a complex mixture of organic pollutants such as lignin, fatty acids, volatile organic compounds, and phenols and inorganic contaminants including sulfates, nitrates, chlorides, and toxic heavy metals like lead, cadmium, and mercury (Haile *et al.* 2021; Kansal *et al.* 2011).

The growing global demand for paper products, driven by expansion in packaging, education, e-commerce, and hygiene sectors, has further intensified this environmental challenge. At the same time, stringent environmental regulations and tighter discharge standards have exposed the limitations of conventional wastewater treatment technologies (Karthik *et al.* 2022). Traditional treatment approaches including sedimentation, activated sludge processes, coagulation, and filtration often fail to remove persistent pollutants such as lignin, chlorinated organic compounds, and heavy metals. These contaminants are persistent, toxic, and resistant to biological degradation, thereby posing long-term ecological and human health risks (Patel *et al.* 2021; Vidal *et al.* 2021).

1.1 Background of Pulp and Paper Industry Wastewater

Wastewater generated from pulp and paper mills typically exhibits high chemical oxygen demand (COD: 1000–7000 mg/L) and biological oxygen demand (BOD: 300–1500 mg/L) due to the presence of persistent organic

matter, particularly lignin. Lignin is a highly aromatic, branched phenylpropane polymer that shows strong resistance to microbial hydrolysis and depolymerization, resulting in dark-colored and toxic effluents (Jagaba *et al.* 2023; Li *et al.* 2024). Chlorinated organic compounds formed during bleaching operations such as dioxins and furans are of particular concern due to their carcinogenic nature. In addition, heavy metals introduced through pulping chemicals, additives, and equipment corrosion tend to bioaccumulate in aquatic organisms, ultimately transferring through the food chain and posing severe risks to both ecosystems and human health (Kumar *et al.* 2021a).

Conventional biological and physico-chemical treatment methods often exhibit limited efficiency in removing these contaminants, primarily due to incomplete degradation, secondary sludge generation, and high operational costs. Even after treatment, the effluent frequently fails to comply with global discharge standards, highlighting a critical technological gap and the urgent need for more effective and sustainable treatment solutions.

1.2 Emerging Technologies

To address these limitations, advanced wastewater treatment technologies, including advanced oxidation processes (AOPs), membrane filtration, and adsorption-based methods have been extensively investigated. Although these technologies are well-established, recent research has focused on improving their performance through novel materials, optimized operational conditions, and hybrid system configurations. Advanced oxidation processes rely on the generation of highly reactive hydroxyl radicals to degrade persistent pollutants, but their application is often constrained by high energy demand, chemical consumption, and the formation of potentially toxic by-products (Giannakis *et al.* 2021). Membrane filtration offers excellent separation efficiency, yet its widespread adoption are limited by membrane fouling, frequent replacement requirements, and high capital costs. Among these approaches, adsorption-based technologies particularly those employing advanced nanomaterials have gained considerable attention due to their simplicity, versatility, and eco-efficiency (Kolya *et al.* 2023). Carbon-based nanomaterials and nanocellulose, especially graphene oxide (GO), have emerged as promising candidates for wastewater remediation as illustrated in Fig. 1 (Komal *et al.* 2024). These materials exhibit high pollutant affinity, rapid adsorption kinetics, and good regeneration potential. However, systematic evaluations of their application to pulp and paper mill wastewater especially complex effluents containing lignin, dyes, and chlorinated organics remain limited (Gupta and Shukla, 2020).

1.3 Environmental Impact and Treatment Challenges

Untreated or partially treated pulp and paper effluents cause severe ecological and public health impacts. High COD and BOD levels deplete dissolved oxygen in receiving water bodies, leading to hypoxic conditions and large-scale aquatic mortality. Dioxins and heavy metals bioaccumulate and disrupt endocrine systems in wildlife and humans (Singh *et al.* 2023). Nutrient-rich compounds such as nitrates and phosphates accelerate eutrophication, further degrading water quality, as illustrated in Fig. 2 (Kashif *et al.* 2019).

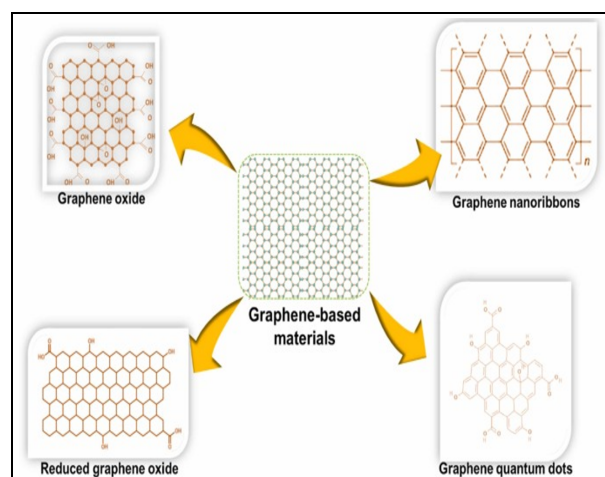


Fig. 1: The graphene-based materials covered in the subsequent sections (Komal *et al.* 2024)

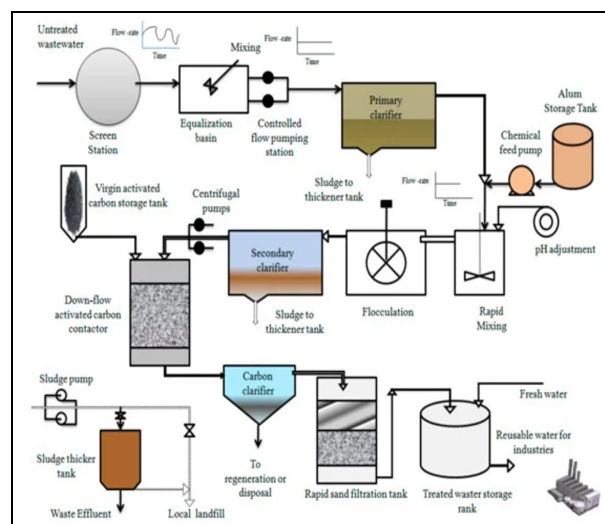


Fig. 2: Treatment process flow diagram based on proposed treatment technology (Kashif *et al.* 2019).

Water reuse and resource recovery from treated pulp and paper wastewater remain limited, as conventional systems are generally unable to recover clean water or valuable by-products efficiently (Lekavičius *et al.* 2021). This limitation highlights the importance of advanced materials and hybrid treatment

technologies capable of removing multiple pollutants while enabling reuse, recovery, and compliance with zero-liquid-discharge policies (Fig. 3).

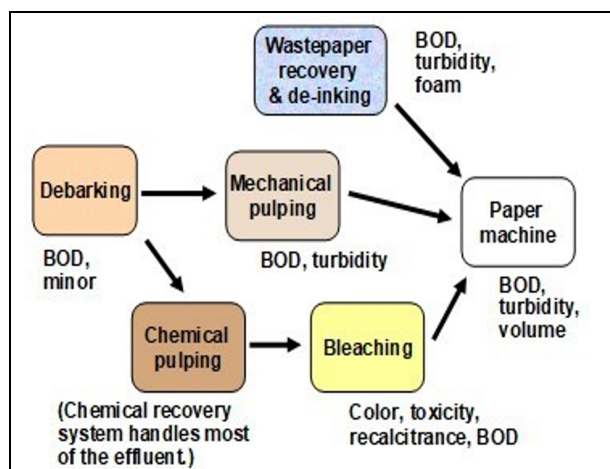


Fig. 3: Types of contaminated water generated in a typical pulp and paper manufacturing plant (Hubbbe *et al.*2016)

1.4 Role of Graphene Oxide in Advanced Wastewater Treatment

Graphene oxide (GO), owing to its two-dimensional structure, large surface area ($\sim 2630 \text{ m}^2/\text{g}$), and abundant oxygen-containing functional groups, has emerged as a versatile adsorbent and catalytic platform for industrial wastewater treatment. It operates through multiple mechanisms, including electrostatic attraction, π - π stacking, and hydrogen bonding, making it effective for removing both organic pollutants (e.g., dyes and lignin) and inorganic contaminants (e.g., heavy metals) (Xiao *et al.* 2023). Its hydrophilicity and surface tunability allow stable dispersion in aqueous media and facilitate integration into hybrid treatment systems.

Functionalization with amine groups, sulfonic acids, or metal oxides further enhances GO's selectivity and adsorption efficiency toward targeted pollutants (Nobre *et al.* 2022). Reduced graphene oxide (rGO) and GO-polymer composites demonstrate improved mechanical strength, regeneration ability, and photocatalytic activity, thereby addressing some scalability and durability challenges associated with conventional nanomaterials (Sharma and Singh, 2021a).

1.5 Objectives and Scope of the Review

This review aims to address critical gaps in the understanding and application of graphene-derived materials for pulp and paper wastewater treatment. The specific objectives are:

- To critically analyze pollutant-specific removal mechanisms of GO and its derivatives, particularly for lignin, heavy metals, and chlorinated organic compounds that are resistant to conventional treatment.
- To evaluate the comparative performance of GO-based adsorbents relative to traditional and emerging technologies, using parameters such as adsorption capacity, kinetics, isotherm models, and reusability.
- To identify operational, environmental, and scalability limitations, including synthesis challenges, regeneration efficiency, toxicity concerns, and barriers to industrial adoption.
- To propose future research directions emphasizing green synthesis, multifunctional composite development, pilot-scale validation, and integration with biological or membrane-based systems for holistic wastewater remediation.

2. METHODOLOGY

2.1 Sources of Literature and Search Strategy

To ensure comprehensive coverage, literature was sourced from five globally recognized scientific databases: Scopus, Web of Science (WoS), Science Direct, Springer Link, and Google Scholar. These platforms were selected for their peer-reviewed content, interdisciplinary coverage, and robust search capabilities. Google Scholar was used strategically to access relevant conference proceedings, grey literature, and recent preprints.

A Boolean search strategy was implemented using combinations of targeted keywords. These included: “pulp and paper wastewater”, “graphene oxide”, “reduced graphene oxide”, “GO-based nanocomposites”, “lignin removal”, “color removal”, “adsorption mechanisms”, “membrane filtration”, “advanced oxidation”, and “wastewater treatment using nanomaterials”.

The search was refined using filters such as language (English only), document type (articles and reviews), and publication period (2015–2024), with greater emphasis placed on studies from the last five

years to ensure the inclusion of the most recent advancements.

2.2 Inclusion Criteria

The review included peer-reviewed studies focused on adsorption-based treatment of pulp and paper industry wastewater, particularly those using graphene oxide (GO), reduced GO, or GO-based composites. Preference was given to articles presenting quantitative data on adsorption performance (e.g., capacity, efficiency, regeneration) and those comparing GO materials with traditional adsorbents like activated carbon or biochar. Due to the limited availability of studies directly employing real pulp and paper mill wastewater, this review also includes GO-based adsorption studies targeting chemically analogous pollutants such as lignin, phenolics, dyes, COD, AOX, and heavy metals, which are characteristic constituents of pulp and paper effluents.

2.3 Exclusion Criteria

Studies were excluded if they lacked relevance to pulp and paper wastewater, did not involve graphene-based materials, or focused solely on non-adsorptive methods. Non-peer-reviewed works, incomplete studies, and publications without sufficient experimental detail were also omitted to ensure scientific rigor and thematic alignment.

3. RESULTS

3.1 Pulp and Paper Industry: Pollution Profile and Environmental Impact

The pulp and paper (P&P) industry is one of the largest industrial consumers of water and a significant contributor to global wastewater generation. As of recent estimates, India ranks among the top paper-producing nations, with rapid industrial expansion contributing to elevated effluent discharge volumes (Del Rio *et al.* 2022). On average, a ton of paper production consumes approximately 60–100 m³ of water, generating wastewater laden with a complex mixture of organic and inorganic pollutants (Patova *et al.* 2021). Despite improvements in closed-loop water systems, many mid- and small-scale paper mills in developing countries still operate with outdated infrastructure, resulting in effluent streams that are variably treated and often discharged without meeting environmental standards (Anegbe *et al.* 2024).

3.2 Wastewater Streams and Contaminants

Pulp and paper mill wastewater is heterogeneous in composition and originates from multiple process points, including wood preparation, pulping (chemical/mechanical), bleaching, and papermaking. Table 1 outlines the typical pollutants, concentration ranges, and their environmental relevance for major mill operations.

Table 1. Characteristics of wastewater from pulp and paper mill operations: major pollutants, concentration ranges, environmental impact, and discharge standards

Wastewater Source/Process	Major Pollutants	Concentration Range (mg/L)	Environmental Impact	Discharge Standards	Reference
Kraft Pulping	COD, BOD, Lignin, Phenolics	COD: 3000–7000; BOD: 1200–3500	Acute aquatic toxicity, oxygen depletion	COD < 250 mg/L	(Kumar <i>et al.</i> 2020; Gholami <i>et al.</i> 2022)
Bleaching Effluent	AOX, chlorinated organics, color	AOX: 15–50; Color > 1000 Pt-Co	Persistent pollutants, bioaccumulation	AOX < 1 mg/L	(Giannakis <i>et al.</i> 2021)
Paper Machine White Water	TSS, Dyes, Fillers, Surfactants	TSS: 800–1500	Aesthetic + chemical contamination	TSS < 100 mg/L	(Lavania <i>et al.</i> 2023); Liu <i>et al.</i> 2021)
Combined Mill Effluent	COD, BOD, TSS, Lignin	COD: 2000–5000; BOD: 800–2000	Eutrophication, high sludge load	BOD < 30 mg/L	(Chowdhary and Mani 2021; Gupta and Shukla 2020; Singh <i>et al.</i> 2023)

This wide range of effluent profiles necessitates multi-stage and highly selective treatment technologies. Traditional biological treatment units often fail to remove complex pollutants like lignin, tannins, and chlorinated derivatives effectively (Sharma *et al.* 2021b).

3.3 Environmental Impacts and Regulation Gaps

The release of untreated or inadequately treated P&P wastewater results in critical ecological impacts. Elevated COD and BOD lead to oxygen depletion in aquatic bodies, negatively impacting fish and benthic organisms (Mandal *et al.* 2023). Chlorinated organics and adsorbable organic halides (AOX) show persistent, bioaccumulative, and toxic (PBT) behaviour, which further distorts aquatic bodies by causing endocrine disruption and mutagenicity (Kumar *et al.* 2020). In addition, high suspended solids and phenolics add turbidity and toxicity to make downstream water unsuitable for reuse or natural body discharge (Goycochea *et al.* 2023). Notwithstanding the availability of regulatory standards such as AOX < 1 mg/L and BOD < 30 mg/L, their enforcement varies, especially in low- and middle-income nations (Carvalho *et al.* 2020).

Life cycle analyses also showed that current treatment options for tertiary treatment are either energy-intensive or economically infeasible at full capacity (Mainardis *et al.* 2024). The need for more sustainable, regenerative, and pollutant-specific solutions is urgent.

3.4 Focus on Lignin: A Persistent Challenge

Among all pollutants, lignin remains the most persistent due to its complex, cross-linked aromatic structure. Lignin resists microbial degradation and persists even after conventional biological treatment (Miao *et al.* 2022; Li *et al.* 2024). It is a primary contributor to the dark coloration of effluents, inhibiting sunlight penetration in aquatic ecosystems and impairing photosynthesis.

Recent studies have emphasized lignin's role as not just a coloring agent but a toxic compound capable of interacting with heavy metals and organic pollutants, forming complex molecules resistant to degradation (Sharma *et al.* 2021c). Furthermore, lignin interferes with downstream disinfection processes and increases the toxicity of treated water when combined with chlorine-based oxidants.

Graphene-based materials due to their affinity for aromatic compounds *via* π - π interactions and large surface areas hold promise for lignin removal. These materials have been shown to outperform activated carbon in adsorption kinetics and capacity under

optimized pH and contact time conditions (Zhang *et al.* 2024).

4. CONVENTIONAL TREATMENT METHODS FOR PULP AND PAPER WASTEWATER

4.1 Traditional Treatment Techniques

The pulp and paper industry employs a variety of conventional treatment techniques to manage its complex and high-load effluents, which are rich in lignin, chlorinated organics, colorants, COD, and suspended solids (Tara *et al.* 2020). The primary goal of these techniques is to reduce pollutant concentrations to acceptable discharge levels; however, they often fall short in completely removing persistent organics like lignin or synthetic dyes (Ribeiro *et al.* 2022). Table 2 below summarizes these traditional methods, outlining their operating mechanisms, efficiencies, and inherent limitations.

4.2 Technical and Operational Limitations

Despite their widespread application, these techniques present significant limitations in addressing the complex and persistent pollutants that characterize pulp and paper mill wastewater. For instance, primary clarification primarily removes suspended solids and is ineffective against dissolved organic compounds. Biological treatments, such as bio-trickling filters and aerated lagoons, perform reasonably well for BOD and ammonia but struggle with persistent compounds like lignin, chlorinated organics, and synthetic dyes (Ribeiro *et al.* 2022).

Chemical coagulation while efficient in color and lignin removal, relies heavily on ferric and alum salts, which leads to substantial sludge production and secondary waste management issues. Moreover, its efficacy is highly pH-dependent, making the process sensitive to influent fluctuations (Kolya and Kang, 2023). Advanced oxidation processes like ozonation offer high removal efficiencies for COD and phenols but are energy-intensive and involve complicated operational setups for ozone generation and handling (Amacosta *et al.* 2024).

Constructed wetlands and anaerobic reactors such as UASB offer more sustainable and energy-efficient alternatives. However, these systems are prone to seasonal performance variability and are often limited in their hydraulic retention capacities, which restricts their use in high-throughput industrial operations (Obaideen *et al.* 2022; Gaur *et al.* 2022). Meanwhile, electrocoagulation, though effective, poses concerns related to electrode maintenance and sludge disposal (Hämäläinen *et al.* 2022).

Table 2. Traditional treatment techniques for pulp and paper wastewater

Treatment Method	Mechanism / Principle	Target Pollutants	Removal Efficiency (%)	Advantages	Limitations	Reference
Primary Clarification	Physical settling by gravity	TSS, coarse solids	40–65%	Simple, low-cost pre-treatment	Does not remove dissolved organics	(Satyam and Patra, 2024a)
Bio-Trickling Filter	Microbial biofilm oxidation	BOD, ammonia, volatile organics	70–80%	Stable performance, low energy	Limited to biodegradable contaminants	(Islam <i>et al.</i> 2023)
Chemical Precipitation	Coagulation using alum/ferric salts	Color, lignin, phosphorus	75–90%	Removes persistent compounds	High sludge production, pH-sensitive	(Kolya and Kang, 2023)
Aerated Lagoon	Extended biological treatment in ponds	BOD, COD, TSS	60–80%	Low cost, large scale	Land-intensive, seasonal performance	(Donkadokula <i>et al.</i> 2020)
Constructed Wetland	Phyto-remediation and sedimentation	TSS, nutrients, color	55–75%	Eco-friendly, low maintenance	Seasonal variation, low hydraulic load	(Obaideen <i>et al.</i> 2022)
Advanced Oxidation (Ozonation)	Free radical degradation	COD, phenols, color	80–95%	Effective for non-biodegradable	Costly gas handling, maintenance	(Giannakis <i>et al.</i> 2021)
Upflow Anaerobic Sludge Blanket (UASB)	Anaerobic digestion under upflow conditions	COD, volatile fatty acids	70–85%	Energy recovery, compact	Long start-up, temperature sensitivity	(Gaur <i>et al.</i> 2022)
Electrocoagulation	Electrochemical destabilization & floc formation	COD, color, TSS	85–95%	High efficiency, lower chemical use	Sludge disposal challenges	(Kumar and Sharma 2022); Gong <i>et al.</i> 2022)
Sustainable Hybrid Filtration	Integrated adsorption & membrane filtration	Heavy metals, COD, dyes	80–90%	Combines multiple mechanisms, environmentally conscious	High material cost, early-stage adoption	(Onyedikachi and Mukah 2024); Yaqoob <i>et al.</i> 2023)

4.3 Emerging Hybrid Techniques: Potential but Underdeveloped

Recent studies have explored sustainable hybrid systems, which integrate adsorption and membrane filtration to simultaneously remove COD, dyes, and heavy metals. These systems exploit the synergistic benefits of multiple removal mechanisms, offering a pathway toward more selective and efficient treatment (Elgarahy *et al.* 2021). However, challenges persist in terms of material costs, fouling resistance, and scale-up feasibility, particularly for continuous industrial application (Gong *et al.* 2022).

4.4 The Need for Advanced Adsorbents

The evident limitation of traditional approaches especially their inefficacy in removing non-biodegradable and toxic organics like lignin and AOX underscore the urgent need for next-generation adsorbents. Materials with high surface area, tunable functional groups, and strong pollutant selectivity are necessary to address the treatment gaps posed by complex pulp and paper effluents. This gap has catalyzed

significant interest in graphene-based materials, known for their high adsorption capacity, reusability, and multi-functionality in targeting a broad range of pollutants.

5. GRAPHENE-BASED ADSORBENTS IN PULP AND PAPER WASTEWATER TREATMENT

Graphene-based adsorbents including pristine graphene oxide (GO), reduced GO (rGO), and a wide variety of functionalized and composite forms have emerged as transformative materials for the remediation of pulp and paper industry effluents. This section synthesizes findings from recent studies captured in Table 3, presenting an in-depth analysis of the materials, target pollutants, adsorption performance, and mechanisms underpinning their effectiveness. It should be noted that direct experimental studies using real pulp and paper mill wastewater are relatively scarce in the literature. Therefore, the studies summarized in Table 3 include GO-based adsorption investigations targeting key pollutant classes such as lignin, dyes, phenolics, COD, chlorinated organics, and heavy metals that are structurally and chemically representative of pulp and paper industry effluents

Table 3: Graphene oxide (GO) & derivatives in pulp and paper wastewater treatment

Go/Derivative Type	Synthesis Method / Modification	Target Pollutants	Adsorption/ Removal Efficiency (%)	Optimum Conditions (pH, Temp, Dosage, Contact Time)	Adsorption Capacity (mg/g)	Adsorption Isotherm / Kinetics Model	Mechanism	Reusability / Regeneration Cycles	Reference
Graphene Oxide (GO)	Modified Hummers' method	Lignin	94%	pH 6.5, 60 min, 0.5 g/L	150.2	Langmuir, Pseudo-second-order	π - π interaction, H-bonding	3–5 cycles with minimal efficiency loss	(Haq <i>et al.</i> 2020)
GO-TiO ₂ Composite	Sol-gel method	Dyes, Lignin	96%	pH 7, 45 min, UV-assisted	160.5	Langmuir	Photocatalysis, electrostatic interaction	Not reported	(Anegbe <i>et al.</i> 2024)
GO-based Membrane	Interfacial polymerization	Color, COD	>90%	Cross-flow filtration	Not specified	—	Size exclusion, surface charge interaction	Not reported	(Jia <i>et al.</i> 2022)
Amine-functionalized GO	Amine grafting	Heavy Metals (Pb ²⁺ , Cd ²⁺)	92%	pH 5–6, 90 min	210	Langmuir, Freundlich	Chelation, electrostatic attraction	Up to 4 cycles	(Velusamy <i>et al.</i> 2021)
Magnetic GO	GO-Fe ₃ O ₄ via co-precipitation	Hg ²⁺ , colorants	88–93%	pH 6, 30 min	145	Pseudo-second-order	Electrostatic, magnetic separation	3 cycles with regeneration	(Zunita, 2021)
GO-Biochar Composite	Biochar doping with GO	Lignin, phenols	85–90%	pH 7, 60 min	120	Freundlich	π - π interaction, surface complexation	Not reported	(Ng <i>et al.</i> 2021)
GO-Polymer Hybrid	Polyamide-GO lamination	COD, organic dyes	90%	Pressure-driven membrane	Not reported	—	Membrane filtration, H-bonding	Not reported	(Bano <i>et al.</i> 2015)
Reduced Graphene Oxide (rGO)	Thermal reduction	Lignin, dioxins	92%	pH 6, 50 min	135	Langmuir	π - π stacking, Van der Waals forces	Not reported	(Kumar <i>et al.</i> 2021b)
GO-ZnO Hybrid	Solvothermal assembly	Dyes, lignosulfonates	97%	Visible light, pH 6.8	168	Langmuir	Photocatalysis, adsorption synergy	2–3 cycles	(Demirel and Rosen, 2023)
Pristine GO	Modified Hummers'	Chlorinated organics	89%	pH 5.5, 40 min	110	Freundlich	Electrostatic, π - π interaction	Not reported	(Esmaceli <i>et al.</i> 2023)
GO-Nanocellulose Hybrid	Physical blending	Dyes, tannins	91%	pH 6–7, 60 min	132	Pseudo-second-order	H-bonding, hydrophobic interaction	4 cycles	(Liang <i>et al.</i> 2023)
GO-AOP System	GO as catalyst support	Phenols, color	95%	UV/H ₂ O ₂ system	Not available	—	Radical oxidation, adsorption	Not reported	(Rayaroth <i>et al.</i> 2023)
GO-Nanocomposite Blend	Functional hybrid nanomaterial	Organic dyes, heavy metals	90–96%	pH 5–7, 30–60 min	~180	Langmuir	π - π stacking, electrostatic interaction	3 cycles	(Satyam and Patra, 2024b)
Functionalized GO Membrane	Layer-by-layer deposition	COD, suspended solids	90–95%	Pressure-driven flow	—	—	Selective barrier, H-bonding	Not reported	(Patel <i>et al.</i> 2021)
GO-PVA Composite	Crosslinked with polyvinyl alcohol	Lignin, dyes	93%	pH 6.5, 40 min	142	Langmuir	π - π stacking, hydrogen bonding	Up to 3 cycles	(Kumar <i>et al.</i> 2020)

5.1 Overview of Graphene Derivatives Used

Various GO derivatives have been synthesized using methods like the Modified Hummers' method (for pristine GO), sol-gel (for GO-TiO₂ composites), solvothermal synthesis (for GO-ZnO hybrids), co-precipitation (for magnetic GO), and physical blending (for GO-nanocellulose hybrids). Functionalization, such as amine grafting and polymer lamination, enhance pollutant specificity and surface interactions. These modified materials significantly outperform traditional adsorbents due to their high surface area, tunable surface chemistry, and capacity to be incorporated into membranes or hybrid systems.

5.2 Target Pollutants and Removal Efficiency

Graphene oxide and its numerous derivatives have shown high promise in addressing the diverse and persistent pollutant load characteristic of pulp and paper

industry wastewater. The pollutant matrix of this effluent is multifaceted predominated by refractory organics such as lignin and phenolic compounds, intense dyes, suspended solids, chlorinated organics, and heavy metals. Recent reviews point out that graphene-based materials possess greater adsorption performance than traditional adsorbents owing to their high surface area, versatility of functional groups, and adjustable physicochemical properties.

5.2.1 Lignin Removal

Lignin is one of the most persistent compounds present in kraft pulping and bleaching wastewater, contributing to color, COD, and toxicity. Pristine GO produced using the modified Hummers' method has been shown to exhibit up to 94% lignin removal efficiency under optimal conditions (pH 6.5, dosage 0.5 g/L, contact time 60 minutes), according to (Haq *et al.* 2020). For example, rGO

which was prepared by thermal treatment, reported upto 92% lignin and dioxin compounds removal (Kumar *et al.* 2021b). The findings validate the tendency of sp^2 hybridized carbon surfaces to bind aromatic macromolecules such as lignin *via* π - π interactions and hydrogen bonding (Gupta *et al.* 2022).

5.2.2 Dyes and Lignosulfonates

Colored compounds including synthetic dyes and lignosulfonates represent a significant problem due to their esthetic and ecotoxicological potential. GO-based nanocomposites have exhibited superior removal efficiencies for such pollutants (Ilyas *et al.* 2022; Goswami *et al.* 2024). For example, Demirel and Rosen (2023) prepared a GO-ZnO hybrid *via* solvothermal processes that showed 97% removal of dyes and lignosulfonates under visible light irradiation. Similarly, Satyam and Patra (2024b) prepared functional GO-nanocomposite blends that attained up to 96% dye removal with broad-spectrum adsorption through electrostatic attraction and π - π stacking mechanisms.

5.2.3 Heavy Metal Sequestration

Heavy metals, although less prevalent than organic pollutants in pulp and paper mill effluents, are often present in trace amounts, especially in integrated industrial sites. Functionalization of GO with amine groups significantly enhances its metal-binding capacity (Zhao *et al.* 2021). Velusamy *et al.* (2021) reported that amine-functionalized GO achieved a maximum adsorption capacity of 210 mg/g for divalent metal ions like Pb^{2+} and Cd^{2+} . The primary adsorption mechanisms include electrostatic attraction and chelation between metal cations and nitrogen-containing functional groups, offering a highly selective and effective approach for metal removal.

5.2.4 Phenolic and Chlorinated Organics

Phenolic compounds and chlorinated organics, commonly generated from the bleaching process, are toxic even at low concentrations and resistant to conventional biological treatment. GO-based systems such as the GO-AOP hybrid (advanced oxidation process) have been shown to achieve up to 95% removal of phenols and color, using radical oxidation pathways under UV/ H_2O_2 systems (Rayaroth *et al.* 2023). Additionally, pristine GO showed 89% efficiency in adsorbing chlorinated organics, driven by electrostatic interactions and π - π stacking (Esmaeeli *et al.* 2023).

5.2.5 Multi Component Wastewater Handling

In complex industrial effluents where multiple pollutant classes coexist, hybrid GO structures such as GO-nanocellulose blends (Liang *et al.* 2023) and GO-polymer membranes (Bano *et al.* 2015) have demonstrated broad-spectrum pollutant uptake,

combining mechanisms like surface complexation, membrane filtration, and hydrophobic interactions. These materials present a multifunctional solution for treating real wastewater samples with COD, color, suspended solids, and organics simultaneously (Worku *et al.* 2023).

5.3 Optimal Operating Conditions and Adsorption Models

Optimizing operating parameters is vital to enhance the adsorption efficiency of GO-based materials for pulp and paper wastewater treatment. Throughout the studies, pH was identified as a key factor determining GO surface charge and ionization status of pollutants. For the majority of the GO composites, the best adsorption efficiency was invariably found to occur within the pH range of 5.5 to 7.0, which is optimal for slightly acidic to neutral conditions. This pH range maximizes π - π interactions with aromatic substances such as lignin and sustains effective electrostatic attraction toward metal ions and dye molecules.

Contact time is also a critical factor, with the equilibrium usually obtained in 30 to 60 minutes for the majority of GO derivatives. For example, pure GO and rGO attained more than 90% pollutant removal within 40–60 minutes (Haq *et al.* 2020; Kumar *et al.* 2021b), whereas magnetic GO composites exhibited even more rapid kinetics because of synergistic separation and surface activity (Zunita, 2021). Adsorbent dose, usually between 0.3–0.5 g/L, was sufficient for sustaining high surface area and providing enough adsorption sites.

5.4 Mechanistic Insights into GO-based Adsorption

The high adsorption efficiency of GO and its derivatives is underpinned by their multifunctional surface chemistry and structural adaptability. π - π stacking interactions play a dominant role in capturing aromatic molecules such as lignin, dyes, and polycyclic organics. These interactions are facilitated by the delocalized π -electron system of graphene sheets and the conjugated structures of the pollutants, forming stable complexes on the GO surface.

Hydrogen bonding is another critical mechanism, especially for phenolic and hydroxyl-containing pollutants, where oxygenated functional groups ($-OH$, $-COOH$, $-C=O$) on GO establish strong intermolecular bonds. Likewise, electrostatic attraction contributes significantly to the adsorption of charged species like metal ions (e.g., Pb^{2+} , Cd^{2+} , Hg^{2+}) and anionic dyes. Functionalization techniques such as amine grafting and polymer blending further enhance this mechanism by introducing electron-donating or protonated groups.

Moreover, in hybrid systems, additional pathways contribute to pollutant removal. For example, GO-TiO₂ composites exhibit photocatalytic degradation under UV or visible light, breaking down dyes and organics into smaller, less toxic compounds (Demirel and Rosen, 2023). GO-based membranes and polymer-GO laminates introduce size exclusion and charge-based filtration, making them ideal for suspended solids and high molecular weight pollutants.

5.5 Reusability and Regeneration Potential

A key factor influencing the practical applicability and economic viability of GO-based adsorbents is their reusability across multiple adsorption-desorption cycles. Encouragingly, many studies report minimal efficiency loss over 3 to 5 regeneration cycles, particularly for GO-PVA composites, amine-functionalized GO, and GO-nanocellulose hybrids (Velusamy *et al.* 2021; Liang *et al.* 2023). These materials retained over 85% of their original adsorption capacity, indicating structural integrity and surface activity over time.

Regeneration techniques commonly employed include mild acid or alkaline washing, thermal

desorption, and solvent extraction (Chen *et al.* 2022). The relatively mild regeneration conditions required suggest that GO-based adsorbents can be seamlessly integrated into cyclic treatment systems with minimal operational disruption.

6. COMPARATIVE EVALUATION WITH TRADITIONAL ADSORBENTS

6.1 Application of Carbon-based Adsorbents

The search for sustainable and efficient adsorbents has long positioned carbon-based materials at the forefront of pulp and paper wastewater treatment. Traditional carbonaceous adsorbents such as activated carbon, biochar, clay-based materials, and agricultural waste-derived adsorbents have demonstrated substantial capabilities in pollutant removal due to their high surface area, porosity, and functional groups (Manna and Sen, 2023). These materials are frequently employed to target dyes, color, COD, BOD, phenols, and even heavy metals, which are common in effluents from paper and pulp mills.

Table 4. GO vs traditional adsorbents

Adsorbent Type	Pollutant Removed	Adsorption Capacity (mg/g)	Optimum pH / Conditions	Reusability	Cost-effectiveness	Reference
Activated Carbon	Dyes, COD, Lignin	120–200	pH 5–7, 60–90 min	Moderate (3–5 cycles)	Moderate–High (commercial)	(Zhou <i>et al.</i> 2019)
Polymeric Resins	Dyes, organics	150–250	pH 6–8, 30–60 min	Good (>5 cycles)	Costly synthesis	(Moradi and Sharma, 2021)
Biochar	Dyes, lignin, color	100–180	pH 6–7, 45–60 min	Low to moderate	Low-cost (agro-waste derived)	(Aragaw and Bogale, 2021)
Agricultural Waste (e.g., husk, shells)	Dyes	80–130	pH 4–6, 40–60 min	Limited (2–3 uses)	Very cost-effective	(Bilal <i>et al.</i> 2022)
Clay-based Adsorbents	Dyes, heavy metals	90–140	pH 5.5–7, 30–60 min	Moderate	Affordable, readily available	(Vishnu <i>et al.</i> 2022)
Nanocomposites (non-GO)	Organic dyes, Pb ²⁺ , Cd ²⁺	150–220	pH 6–7, UV-assisted	Good (>4–5 cycles)	Moderate–High	(Akhtar <i>et al.</i> 2024)
Biowaste-derived Adsorbents	Methylene blue, dyes	100–160	pH 6, 60 min	Limited	Excellent for low-cost removal	(Hamad and Idrus 2022)
Traditional Mixed Matrix Adsorbents	Color, BOD, COD	110–160	pH 6–7, 90 min	Moderate	Feasible for industrial use	(Kumar <i>et al.</i> 2021b)

6.2 Performance Landscape of Traditional Adsorbents

As seen in Table 4, activated carbon continues to set the benchmark for dye and lignin removal with adsorption capacities ranging between 120–200 mg/g under optimal pH conditions (5–7) and contact durations of 60–90 minutes (Zhou *et al.* 2021). Its relatively moderate cost and established reusability (up to 5 cycles) make it suitable for industrial deployment, although pore

clogging and regeneration inefficiencies remain a concern. Polymeric resins offer a higher adsorption range (150–250 mg/g) and better regeneration potential (>5 cycles), particularly for organic dye contaminants. However, they are cost-intensive due to complex synthesis processes and are less attractive for large-scale use without cost mitigation strategies.

Biochar and bio waste-derived adsorbents, especially those sourced from agricultural residues, have

gained traction due to their eco-friendly synthesis, low cost, and tunable porosity. For example, biomass-derived adsorbents have shown moderate-to-good removal efficiency for dyes and lignin (100–160 mg/g) under pH 6–7 but are typically limited to 2–3 reuse cycles, making them more suitable for single-use or batch treatment system (Zhao *et al.* 2021). Clay-based adsorbents provide a cost-effective solution for treating heavy metals and dyes. They perform moderately well (90–140 mg/g) but suffer from slower kinetics and structural instability at low pH, limiting their universal applicability (Vishnu *et al.* 2022).

Recent developments in nanocomposites have also shown promise, with improved adsorption capacities (~220 mg/g) and enhanced surface interactions under UV-assisted or catalytically active conditions. These systems strike a better balance between performance and regeneration but still incur higher costs due to specialized nanomaterial synthesis (Xiang *et al.* 2022).

6.3 Critical Challenges with Traditional Adsorbents

Traditional adsorbents like activated carbon, biochar, and agricultural waste-based materials struggle to effectively remove persistent pollutants such as lignin, chlorinated organics, and synthetic dyes. Their regeneration efficiency is generally low, especially in biochar and agro-residue-based adsorbents, and they often require high energy or chemical input for reuse. Additionally, instability under fluctuating pH or ionic strength and the tendency for fine particle leaching reduce their long-term applicability. As Kumar *et al.* (2021b) highlighted, mixed matrix adsorbents possess average adsorption capacity (110–160 mg/g) but not the selectivity and operational stability required in actual-time wastewater treatment systems.

6.4 Rationale for Transitioning to Graphene-Based Adsorbents

Graphene-based adsorbents, particularly GO and its composites, have considerable merits over conventional materials. Their very high surface area (~2630 m²/g), rich oxygen-containing functional groups, and layered nanostructure exhibit strong interaction sites for a broad scope of pollutants, such as dyes, lignin, and heavy metals. Unlike conventional adsorbents, GO exhibits superior performance in variable pH environments and allows for facile surface modification to target specific contaminants. Studies have shown GO and its hybrids achieve higher adsorption capacities (often exceeding 150–200 mg/g) with rapid kinetics and better regeneration potential across multiple cycles. These properties are critical for addressing the complex, high-strength effluent characteristics typical of pulp and paper mill wastewater. Furthermore, the ability to

integrate GO into membranes or hybrid systems enhances its practical utility for scalable, continuous-flow operations, establishing it as a next-generation material for sustainable and efficient industrial wastewater treatment.

7. MAPPING LITERATURE INSIGHTS TO TREATMENT OPPORTUNITIES

A comparative examination of recent GO-based technologies reveals their strategic applicability to distinct classes of pollutants in pulp and paper wastewater. For instance, pristine GO has shown promise in facilitating pollutant-specific interactions through its oxygen-containing groups, although its experimental validation in real wastewater remains underexplored (Nebol'Sin *et al.* 2020). Polymer-functionalized GO hybrids demonstrated efficient removal of dyes and organics, but limited research exists on their performance in lignin-rich matrices (Nicosia, 2021) as shown in Table 5. To address this, lignin-derived porous carbons, structurally similar to GO, offer excellent dye removal and recyclability, paving the way for GO-lignin hybrid adsorbents (Zhu *et al.* 2022). Sustainable bio sorbents, such as lignin-based GO composites, have yielded high adsorption capacity but require further studies on regeneration (Zhang *et al.* 2024). Notably, integrating GO into microbial fuel cells and photocatalysts (e.g., BiVO₂-GO, Cu₂O-Au/GO) has opened dual pathways for energy recovery and pollutant degradation, though scalability remains a challenge (Selvasembian *et al.* 2022; Subhiksha *et al.* 2021; Yu *et al.* 2022). Emerging studies also model greenhouse gas emissions and perform life cycle assessments of GO-integrated tertiary systems, highlighting their broader sustainability potential (Han *et al.* 2024; Mainardis *et al.* 2024). These studies together demonstrate that GO and its derivatives are not just efficient adsorbents, but adaptable, multifunctional platforms capable of addressing targeted treatment goals in complex industrial effluents as shown in Table 6.

8. CRITICAL DISCUSSION AND KNOWLEDGE GAPS

Graphene oxide and its derivatives have demonstrated superior adsorption performance over traditional adsorbents, with removal efficiencies exceeding 90% for lignin, dyes, chlorinated organics, and heavy metals. Functionalized variants such as GO-ZnO hybrids, amine-GO, and GO-polymer membranes provide targeted pollutant affinity, improved kinetics, and regeneration. However, the wide variance in reported adsorption capacities and mechanisms stems from differences in synthesis methods, pollutant characteristics, pH ranges, and experimental designs. Many studies remain restricted to batch-scale trials using synthetic solutions, with insufficient exploration of real, lignin-rich industrial effluents. Regeneration data beyond

3–5 cycles is scarce, and long-term stability in continuous systems is largely unverified. Moreover, while π – π stacking, hydrogen bonding, and electrostatic interactions are frequently cited, a unified mechanistic framework is lacking. There is also a dearth of toxicological studies on GO leachates, especially under electrochemical or photocatalytic applications. These gaps underscore the need for holistic evaluations involving mixed pollutant scenarios and real-time operational stressors.

9. FUTURE RESEARCH DIRECTIONS

To address existing gaps, future research must prioritize development of advanced hybrid materials such as GO-MOFs, GO-biochar composites, and green-synthesized GO from agro-wastes. These materials should integrate adsorption with photocatalysis, membrane separation, or electrochemical functions for

enhanced multi-pollutant remediation. Emphasis should be placed on long-term performance, regeneration efficiency, and structural stability under variable industrial conditions. Pilot-scale trials and continuous-flow reactors using actual pulp and paper effluents are essential to validate lab-scale results. Moreover, toxicity assessments especially ecological impacts and microbial tolerance must be systematically incorporated. Life cycle assessment, techno-economic analysis, and carbon footprint analysis may provide understanding regarding the sustainability of GO-based systems. Such innovations in line with circular economy principles such as lignin sludge valorization or biomass processing-derived supports will ensure regulatory clearance and industrial uptake. Last but not least, integration into interdisciplinary frameworks based on material science, environmental engineering, and policy integration can expedite GO technology transfer from laboratory to land.

Table 5. Global research trends & research gaps

Go Material / Composite Used	Pollutants Studied	Key Findings	Identified Limitations	Suggested Future Research	Reference
Pristine GO	Interaction with oxygen groups	Elucidated complex multistage oxygen interactions on GO, useful for adsorptive tuning	Lacked experimental wastewater validation	Explore pollutant-specific interactions in real wastewater matrices	(Nebol'Sin <i>et al.</i> 2020)
Polymer-functionalized GO	Dyes, organics	Highlighted versatility of polymer-GO hybrids	Limited data on pulp/paper matrices	Apply polymer-GO to lignin-rich effluents	(Nicosia, 2021)
Lignin-derived porous carbon (GO-like)	Organic dyes	Demonstrated rapid dye adsorption and recyclability	Not GO-composite; comparative GO data missing	Combine lignin-porous carbon with GO for hybrid adsorbents	(Zhu <i>et al.</i> 2022)
Lignin-based GO biosorbents	Mixed pollutants	Emphasized sustainable biosorbent synthesis with high capacity	No long-term stability tests	Focus on regeneration and scaling of biosorbents	(Zhang <i>et al.</i> 2024)
Porous polyamine lignin microsphere (lignin-based adsorbent, potential for GO-composite development)	COD, color, organic pollutants	High adsorption efficiency and stability of lignin-derived microspheres for pollutant removal	Limited to batch-scale studies; no integration with GO or hybrid systems	Combine lignin microspheres with GO-based materials for synergistic adsorption	(Pang <i>et al.</i> 2023)
GO in Microbial Fuel Cells	Heavy metals, COD	Highlighted dual benefit of treatment and energy	Tech untested in pulp/paper scale	Integrate GO-MFC in pilot industrial units	(Selvasembian <i>et al.</i> 2022)
BiVO ₂ -GO photocatalysts	Organic pollutants	Achieved visible-light degradation of persistent organics	No application to lignin/color components	Adapt for lignin and colorant treatment under solar light	(Subhiksha <i>et al.</i> 2021)
Cu ₂ O-Au/GO anchored on wood	Dyes, organic pollutants	Synergistic photodegradation with natural support	Costly catalyst material	Explore cheaper alternatives with same efficiency	(Yu <i>et al.</i> 2022)
GO integrated in biological models	GHG Emissions in WWTP	Developed emission models incorporating GO-based systems	Focused on modeling, not treatment validation	Validate predictive models in pilot GO-biological reactors	(Han <i>et al.</i> 2024)
GO-integrated tertiary systems	COD, color	Compared tertiary treatments using LCA and cost	GO not isolated as variable	Conduct LCA isolating GO membranes vs traditional AOPs	Mainardis <i>et al.</i> 2024)

Table 6. Opportunities and challenges in GO-based treatment

Opportunities	Challenges	Proposed Solutions	Potential For Industrial Scale-Up	Reference
High adsorption capacity of GO for diverse pollutants (dyes, lignin, metals)	Limited large-scale validation and pilot studies	Implement continuous flow and real effluent testing	Requires robust field trials and long-term assessments	(Selvasembian <i>et al.</i> 2022)
Integration of GO with catalytic systems (e.g., Fe-TiO ₂) for enhanced degradation	Stability of composites under harsh wastewater conditions	Surface modification and doping to improve durability	Promising for hybrid adsorbent systems in industry	(Puri and Verma, 2022)
GO-based systems may lower greenhouse gas emissions via reduced sludge production	Lack of data on environmental emissions and life cycle impacts	Incorporate LCA and carbon footprint analysis	Useful in sustainability assessments for scale-up	(Han <i>et al.</i> 2024)
Potential for eco-friendly and cost-effective treatment <i>via</i> GO composites	High initial synthesis cost of GO and derivatives	Explore green synthesis and biomass-derived GO	Moderate to high, contingent on material cost reduction	(Mainardis <i>et al.</i> 2024)
Electrochemical compatibility of GO enables integration into AOPs and EC systems	GO degradation or fouling during electrochemical cycles	Periodic regeneration and advanced coating	Viable with electrode-GO hybrid optimization	(Moradi <i>et al.</i> 2021)
GO can be tuned to capture heavy metals and persistent organics	Toxicity concerns of nanomaterials in biological treatment units	Study eco-toxicity, microbial tolerance, and post-treatment filtration	Must align with environmental regulations and reuse potential	(Sharma <i>et al.</i> 2021d)
Biomass-based flocculants can be combined with GO for synergistic action	Lack of operational frameworks for such hybrid models	Develop protocols for dual-functional GO-biomass systems	Attractive due to circular economy potential	(Sun <i>et al.</i> 2023)
Agro-residues can be used to synthesize GO-membrane composites	Mechanical strength and fouling of hybrid membranes	Reinforce composites with nanocellulose or MOFs	High if mechanical integrity is ensured	(Worku <i>et al.</i> 2023)
GO-MOF and GO-polymer materials show tailored selectivity	Complex synthesis and limited recyclability	Simplify protocols using bio-compatible linkers	High for selective treatment (e.g., lignin dyes)	(Tao <i>et al.</i> 2023)
Recent innovations in eco-friendly pulp wastewater decontamination align with GO strategies	Fragmented research on GO's regulatory and scale-up pathways	Interdisciplinary frameworks linking GO tech with pulp industry needs	Essential for adoption in industrial pulp sectors	(Gupta and Kapoor, 2024)

10. CONCLUSION

This review confirms that GO and its composites outperform conventional adsorbents in treating pulp and paper mill wastewater, particularly for the removal of persistent, industry-specific pollutants such as lignin, color-causing dyes, chlorinated organics, and trace heavy metals. The complex composition of pulp and paper effluents characterized by high COD, dark coloration, aromatic macromolecules, and bioaccumulative contaminants makes GO-based materials especially relevant, as their high surface area, tunable functional groups, and multi-mechanistic adsorption pathways enable effective interaction with lignin-rich and phenolic matrices. Key performance trends identified in pulp and paper-focused studies highlight functionalization (amine-, metal oxide-, and polymer-modified GO), hybridization with membranes or catalytic systems, and integration into tertiary treatment stages as promising strategies for enhancing pollutant removal efficiency and selectivity. These approaches demonstrate improved adsorption kinetics, high removal efficiencies (>90%), and moderate reusability (3–5 cycles) under conditions relevant to

paper mill effluents. However, most reported investigations remain confined to laboratory-scale batch studies using synthetic or partially simulated pulp and paper wastewater, with limited validation using real mill effluents containing mixed pollutants such as lignin, AOX, color, and suspended solids. Uncertainty persists regarding long-term regeneration performance, adsorbent stability under continuous flow, and the environmental safety of GO nanomaterials when applied at industrial scale. These challenges are particularly critical for pulp and paper applications, where large wastewater volumes, variable effluent composition, and strict discharge standards demand robust and scalable solutions. Future research must therefore prioritize pulp and paper-specific pilot-scale demonstrations, continuous-flow operation, and long-term performance evaluation using real effluents. Scalable and green synthesis routes, life-cycle assessment, toxicity evaluation, and alignment with industrial wastewater regulations are essential to support practical deployment. Overall, GO-based technologies represent a highly promising pathway for sustainable and high-efficiency treatment of pulp and paper industry wastewater, but targeted innovation and industry-oriented validation are

required to bridge the gap between laboratory success and full-scale implementation.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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