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A Performance-Based Comparison of Advanced Oxidation Processes, Biochar Adsorption, and Membrane Filtration for Removing Pharmaceuticals from Municipal Wastewater

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Abstract: **Pharmaceutical residues in municipal wastewater pose a critical environmental and public health challenge, as conventional activated sludge treatment removes less than 50% of many pharmaceutical compounds. This systematic review compares three advanced tertiary treatment technologies: advanced oxidation processes (AOPs), biochar adsorption, and membrane filtration. The comparison evaluates removal efficiency, energy consumption, capital and operational costs, technology readiness level, and transformation product risks. Reverse osmosis and photo-Fenton oxidation achieve the highest removal efficiencies, exceeding 95% across diverse pharmaceutical classes. However, they require substantial energy inputs (1.5 to 6.0 and 0.5 to 2.0 kWh/m³, respectively) and significant capital investment. Biochar adsorption offers the most cost-effective solution for moderate removal targets (70 to 90%) with minimal energy requirements (less than 0.1 kWh/m³), making it particularly valuable for resource-limited settings. Hybrid systems that combine oxidation with biological treatment or adsorption consistently outperform standalone technologies by 10 to 20 percentage points. No single technology dominates across all decision criteria. Technology selection should incorporate treatment objectives, regulatory requirements, budget constraints, and local implementation capacity based on specific context. Future municipal wastewater systems will likely use combinations of technologies rather than relying on single universal solutions.**

Keywords: **advanced oxidation processes, biochar, pharmaceutical removal, membrane filtration, municipal wastewater treatment, technology comparison.**

1. INTRODUCTION

1.1 Background: Pharmaceutical Contamination in Municipal Wastewater

Pharmaceuticals enter municipal wastewater through human and veterinary excretion, disposal of unused medications, and discharges from healthcare facilities and manufacturing operations. Municipal secondary effluents typically contain pharmaceutical concentrations ranging from 0.1 to 10 µg/L (Verlicchi et al., 2012), with specific compounds varying according to population demographics, disease prevalence, and seasonal medication patterns. Commonly detected pharmaceuticals include carbamazepine (anticonvulsant), diclofenac (nonsteroidal anti-inflammatory), ibuprofen (analgesic), sulfamethoxazole (antibiotic), atenolol (beta-blocker), and naproxen (NSAID), with concentrations frequently exceeding single-compound and mixture toxicity thresholds in receiving waters (Huber et al., 2003).

Pharmaceutical residues are concerning because of their biological activity and environmental persistence. These compounds are designed for efficacy at low doses and resistance to metabolic degradation. These properties are advantageous in the human body but problematic in aquatic environments. Chronic exposure to pharmaceutical mixtures alters fish reproductive behavior and endocrine function, reduces bacterial fitness, and promotes the dissemination of antibiotic resistance genes in environmental microbial communities (Verlicchi et al., 2012). These effects occur at environmentally relevant concentrations, posing genuine ecological risks.

Regulatory and water security drivers have elevated pharmaceutical control requirements. Potable reuse projects routinely mandate removal of indicator micropollutants including pharmaceuticals to achieve very high log-reduction values (>3–4 orders of magnitude) through multi-barrier treatment and monitoring. The European Union's Watch List mechanism identifies diclofenac, estradiol, and certain antibiotics as substances requiring environmental monitoring (Escher & Fenner, 2011). These regulatory developments have transformed pharmaceutical removal from optional advanced treatment to essential infrastructure for many municipalities.

1.2 Limitations of Conventional Wastewater Treatment

Activated sludge processes were designed to remove bulk organic carbon, nutrients, and pathogens. Pharmaceutical removal in these systems is limited and variable. Readily biodegradable compounds like ibuprofen show 40–99% removal through combined sorption and microbial degradation, while recalcitrant compounds like carbamazepine achieve only 10–30% removal even at extended sludge retention times of 20–30 days (Clara et al., 2005). This performance gap exists because pharmaceuticals are designed for metabolic stability. These characteristics directly oppose microbial degradation.

Extended sludge retention time can improve biodegradation for some compounds but increases aeration energy demand and operational risks such as sludge bulking. Process enhancements designed to improve nutrient removal, such as anoxic–oxic sequencing, have not consistently improved pharmaceutical removal (Verlicchi et al., 2012). The fundamental constraint is that conventional activated sludge was not designed for removal of trace organics. This limitation has created a recognized treatment gap. Regulatory and societal expectations for pharmaceutical control are rising, but legacy treatment infrastructure was not designed to achieve the required level of removal.

1.3 Technology Overview

Three major technology families have emerged as practical tertiary solutions for pharmaceutical control in municipal wastewater.

Advanced Oxidation Processes rely on in situ generation of powerful oxidizing species, most notably hydroxyl radicals, which react with a broad spectrum of organic molecules. Ozonation, Fenton and photo-Fenton oxidation, heterogeneous photocatalysis, and electrochemical oxidation each generate hydroxyl radicals through different mechanisms, resulting in distinct trade-offs between selectivity, energy demand, and operational simplicity (Reungoat et al., 2012). The principal advantage is their ability to transform structurally recalcitrant pharmaceuticals through oxidative degradation.

Biochar Adsorption removes pharmaceuticals by accumulating them on a porous carbonaceous surface via physical and chemical interactions. By selecting feedstock and pyrolysis conditions, the resulting biochar can be tailored to maximize uptake of hydrophobic and aromatic pharmaceuticals (Tan et al., 2016). Unlike oxidative or membrane processes, adsorption transfers the pharmaceutical from water to the adsorbent surface, allowing for potential regeneration and recovery pathways.

Membrane Filtration, particularly nanofiltration and reverse osmosis, provides a physical barrier to pharmaceuticals based on a combination of molecular size exclusion and electrostatic repulsion (Bellona et al., 2004). Nanofiltration can achieve high rejection for charged and moderately sized molecules at relatively moderate pressure, while reverse osmosis provides near-universal rejection of all pharmaceuticals at the cost of higher energy demand.

These three platforms span a performance/cost/energy continuum. Biochar represents a low-cost, low-energy but more selective approach. In contrast, reverse osmosis provides high-performance, energy-intensive, largely non-selective barriers. Figure 1 illustrates how these technologies integrate into centralized and decentralized municipal treatment and reuse architectures.

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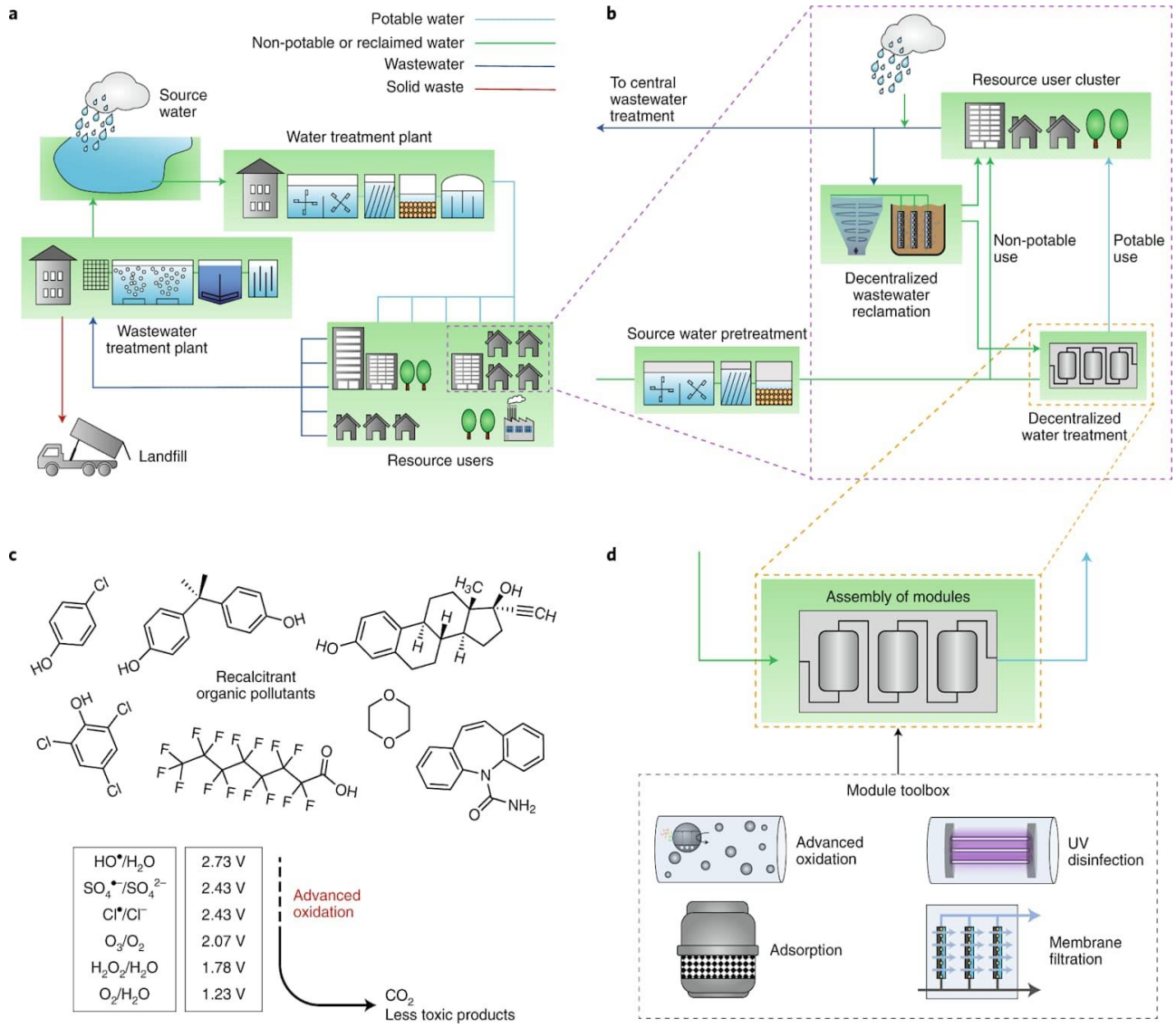

Figure 1. Centralized and decentralized municipal water treatment and reuse architectures, and modular advanced treatment toolbox. Panels (a) and (b) illustrate source water, conventional water and wastewater treatment, and decentralized reuse configurations. Panels (c) and (d) highlight treatment of recalcitrant organic pollutants via advanced oxidation, adsorption, and membrane filtration modules.

1.4 Research Questions and Scope

The primary research question guiding this review is: which of these three technology families offers the most favorable balance of pharmaceutical removal, energy demand, and cost under different operational, regulatory, and economic constraints?

Secondary questions include:

I. How do removal efficiencies compare across technologies for representative pharmaceuticals such as carbamazepine, diclofenac, ibuprofen, and sulfamethoxazole?

II. What are the typical ranges of energy consumption and unit treatment cost associated with each technology?

III. How do transformation products from AOPs affect ecotoxicological outcomes?

IV. Do hybrid configurations combining two or more technologies deliver performance gains relative to single processes?

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2. ADVANCED OXIDATION PROCESSES

2.1 Mechanisms and Process Types

Advanced oxidation processes generate reactive oxygen species that react with pharmaceuticals through oxidative attack. The hydroxyl radical (•OH), an extremely powerful oxidant, is the most important species across all AOPs. It is capable of reacting with most organic molecules.

Ozonation initiates oxidation through direct reaction of the ozone molecule with electron-rich pharmaceutical moieties, including aromatic rings, C=C double bonds, and phenolic structures (Reungoat et al., 2012). This selective direct oxidation is complemented by indirect oxidation where ozone decomposes to generate hydroxyl radicals, especially under alkaline conditions. The selectivity of direct ozonation means that removal efficiency varies across pharmaceutical structures.

Figure 2. Schematic municipal wastewater treatment train showing secondary biological treatment followed by ozone-based advanced oxidation and a polishing biofilm reactor.

Source: Papageorgiou et al., 2020

Fenton and Photo-Fenton Processes generate hydroxyl radicals through reaction of ferrous iron (Fe^{2+}) with hydrogen peroxide (Antonopoulou et al., 2021). In the photo-Fenton variant, UV irradiation regenerates Fe^{2+} from Fe^{3+}, sustaining radical generation and reducing iron consumption (Oller et al., 2011). This regeneration mechanism allows photo-Fenton to treat larger water volumes with lower chemical inputs than dark Fenton.

Heterogeneous Photocatalysis employs a solid photocatalyst, most commonly titanium dioxide (TiO_2), activated by UV or visible light to generate electron/hole pairs (Huber et al., 2003). These drive redox reactions at the catalyst surface, producing hydroxyl radicals that attack pharmaceuticals.

Electrochemical Oxidation applies electrical potential across electrodes, driving oxidation of pharmaceuticals either directly at the electrode surface or indirectly through hydroxyl radical generation (Panizza & Cerisola, 2004). Advanced electrode materials can achieve very high oxidation efficiency but require substantial electrical energy input

Table 1. Advanced Oxidation Process Characteristics

Process Type	Primary Oxidant	Typical Removal Range	Energy Intensity (kWh/m³)	Major Operational Challenge
Ozonation	O_3, •OH (indirect)	60 - 95%	0.05 - 0.30	Ozone transfer; TP formation
Peroxone	Enhanced •OH	75 - 95%	0.10 - 0.40	H_2O_2 supply and cost
Fenton/Dark	•OH	85 - 98%	0.20 - 0.50	pH adjustment; Fe sludge
Photo-Fenton	Enhanced •OH regeneration	90 - 99%	0.50 - 2.00	UV lamp maintenance

Photocatalysis	•OH, $•O_2^-$	50 - 95%	0.30 - 1.50	Catalyst recovery
Electrochemical	•OH, direct oxidation	>99%	20 - 100	Extreme energy cost

2.2 Performance Data and Operational Influence

Pharmaceutical removal by ozonation varies with dose and contact time. In pilot and full-scale studies of municipal secondary effluent, typical removals at doses of 5 to 10 mg O_3/L achieve 60 to 80% for carbamazepine, 80 to 95% for diclofenac and ibuprofen, and 50 to 85% for sulfamethoxazole (Reungoat et al., 2012). Higher ozone doses increase removal but with diminishing returns. Dose/response curves show substantial improvement up to approximately 10 mg O_3/L, after which additional oxidant contributes only modest gains (Hollender et al., 2009).

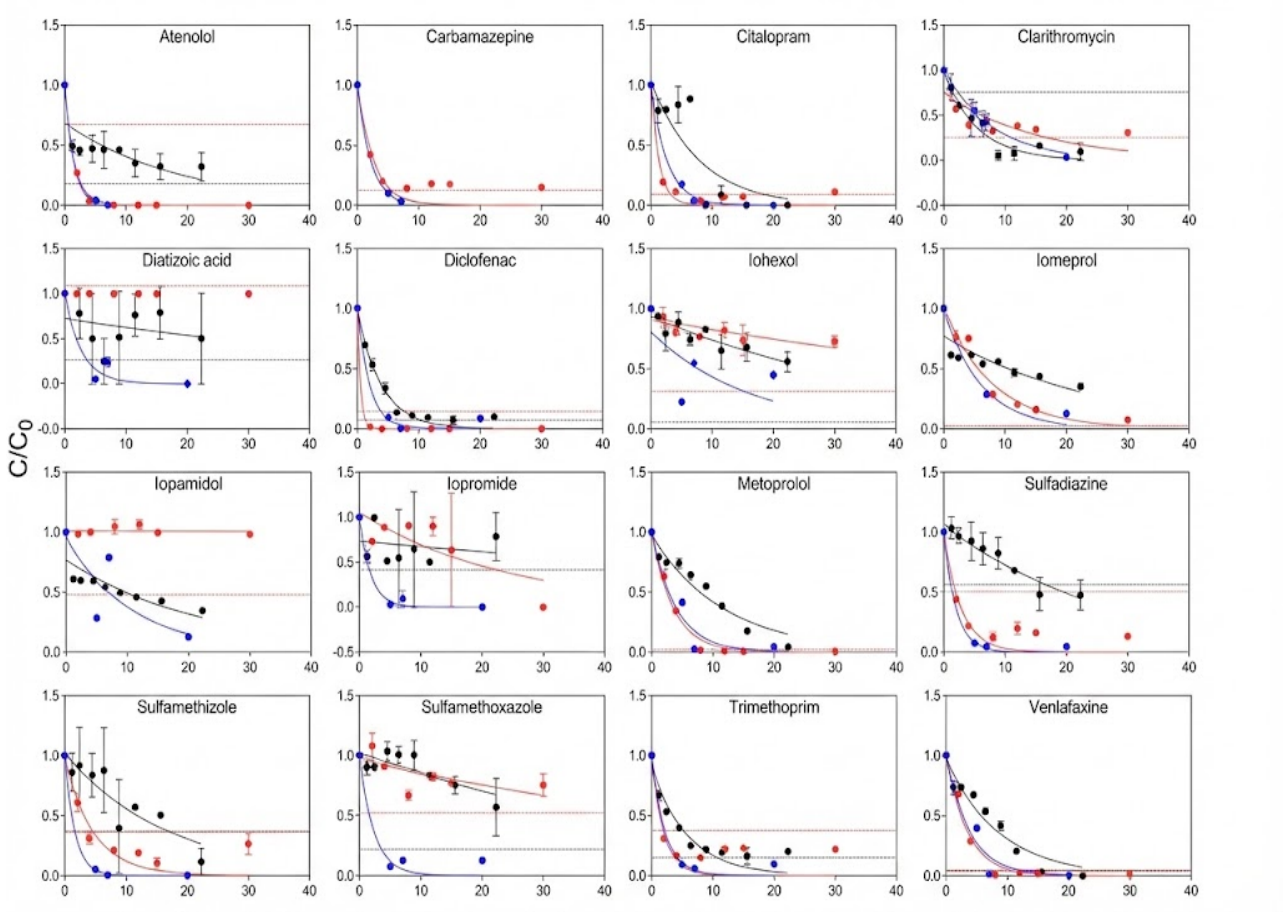

Figure 3. Normalized concentration (C/C_0) of selected pharmaceuticals as a function of delivered ozone dose in pilot and laboratory scale ozonation of municipal wastewater secondary effluent.

Source: Papageorgiou et al., 2020

Fenton and photo-Fenton systems show notably higher and more consistent removal. Dark Fenton typically achieves 85 to 98% removal of most pharmaceuticals within 1 to 4 hours of contact time at Fe^{2+} and H_2O_2 concentrations of 50 to 200 mg/L (Antonopoulou et al., 2021). Photo-Fenton consistently achieves >95% removal for diverse pharmaceutical classes, often reaching >98% mineralization (Oller et al., 2011).

Operational parameters substantially influence performance. Ozonation performs well at neutral to slightly alkaline conditions (pH 7 to 8), though some compounds show enhanced removal at pH 5 to 6 (Reungoat et al., 2012). Fenton processes require acidic pH (2.5 to 4.0) for optimal iron speciation, necessitating chemical adjustment in municipal applications (Antonopoulou et al., 2021). Contact time significantly influences removal. Ozonation reaches approximately

50% of maximum removal within 2 to 5 minutes but typically requires 15 to 30 minutes for 90% removal (Hollender et al., 2009)

2.3 Advantages and Operational Constraints

The principal strength of AOPs is non-selective oxidation across a broad spectrum of pharmaceutical structures and chemical classes. This universality contrasts with adsorption and membrane processes, which show performance variability depending on pharmaceutical properties such as hydrophobicity, charge state, and molecular size (Huber et al., 2003). Fenton and photo-Fenton processes offer the additional advantage of biodegradability enhancement. Partial oxidative breakdown of recalcitrant pharmaceuticals generates intermediate compounds more readily degradable by biological reactors (Oller et al., 2011).

Constraints are substantial. Energy intensity ranges from moderate for ozonation to prohibitive for electrochemical oxidation. For regions with limited electricity supply or high energy costs, this energy demand becomes a decisive barrier. Operational costs ranging from \$0.10 to 0.30/m³ for ozonation to \$1.00 to 3.00/m³ for photo-Fenton create economic pressures in low-income settings (Reungoat et al., 2012). Skilled operation is essential. Ozonation requires off-gas monitoring to prevent atmospheric emissions, while Fenton processes demand precise pH control and automated chemical dosing (Antonopoulou et al., 2021).

2.4 Transformation Products and Ecotoxicity

A significant limitation of AOPs, particularly ozonation, is the formation of transformation products (TPs). These are partial oxidation intermediates that may persist in the environment (Escher & Fenner, 2011). Ozonation breaks pharmaceutical molecules through selective oxidation of reactive functional groups, but often does not achieve complete mineralization (Reungoat et al., 2012). The resulting TPs may accumulate in treated effluent and downstream environments. For example, ozonation of carbamazepine produces epoxide and aldehyde intermediates detected downstream in treated effluent (Hollender et al., 2009).

Research using effect-based bioassays has documented paradoxical outcomes. Treatment removing 60% of a parent pharmaceutical (such as diclofenac) can increase overall estrogenic activity or daphnia immobility in the treated sample relative to the untreated effluent (Escher & Fenner, 2011). This "risk quotient reversal" indicates that TP formation in some ozonation scenarios creates net increases in ecotoxicological effects despite substantial parent compound removal.

Photo-Fenton and electrochemical oxidation minimize TP concerns by driving pharmaceutical transformation toward complete mineralization rather than partial oxidation (Oller et al., 2011). Practical risk management strategies include coupling ozonation with downstream biological treatment or granular activated carbon adsorption. A Swiss wastewater treatment plant upgraded to include post-ozonation, sand filtration, and granular activated carbon reported >95% removal for 12 monitored pharmaceuticals including carbamazepine and diclofenac over continuous 2-year operation (Hollender et al., 2009).

Table 2. Transformation Product Formation and Risk by AOP Type

AOP Type	Typical TP Quantity	TP Ecotoxicity Risk	Risk Quotient Status	Recommended Post-Treatment
Ozonation	Multiple (3 - 8)	Medium - High	Often reverses gains	Biofilter or GAC
Peroxone	Moderate (2 - 4)	Medium	Possible reversal	Secondary treatment
Fenton	Few (1–3)	Low - Medium	Well-controlled	Optional
Photo-Fenton	Minimal (<2)	Low	Favorable mineralization	Not required

Photocatalysis	Variable (2 - 6)	Medium - High	Compound-dependent	Biotreatment recommended
Electrochemical	Minimal	Low	Complete mineralization	Not required

3. BIOCHAR ADSORPTION

3.1 Biochar Characteristics and Production

Biochar is a carbon-rich solid produced by pyrolysis, the thermal decomposition of organic material under oxygen-limited conditions. Pyrolysis typically occurs at temperatures from 300 to 700°C (Yao et al., 2012). The choice of feedstock and pyrolysis temperature jointly determine surface properties. Common feedstocks include agricultural residues, forest biomass, and municipal organic waste streams. Biochar production from waste feedstocks costs substantially less than commercial activated carbon. Typical costs are $100 to 500 per ton compared to $500 to 2000 per ton, making it economically attractive, particularly in regions with abundant biomass (Yao et al., 2012).

Biochar surface area varies from less than 10 m^2/g for material produced at low pyrolysis temperature to greater than 500 m^2/g for activated biochars (Chen et al., 2008). Pyrolysis temperature influences surface chemistry. Higher temperatures (>500°C) increase aromaticity and reduce oxygen-containing functional groups, while lower-temperature pyrolysis (300 to 500°C) retains greater quantities of oxygen groups facilitating polar interactions (Tong et al., 2019).

Activation methods enhance surface properties substantially. Physical activation using steam or CO_2 at elevated temperature creates additional microporosity and can increase surface area 3 to 5 fold relative to non-activated biochar (Yao et al., 2012). Chemical activation using potassium hydroxide, phosphoric acid, or zinc chloride similarly increases surface area while tailoring pore size distribution to match target pharmaceutical dimensions (Chen et al., 2008). The point of zero charge typically ranges from pH 6 to pH 10 depending on oxygen-containing functional group abundance (Ndoun et al., 2021).

3.2 Adsorption Mechanisms

Pharmaceutical uptake by biochar operates through multiple concurrent mechanisms. π-π stacking interactions occur between aromatic rings in pharmaceutical molecules and the graphene-like sheets comprising the biochar matrix (Chen et al., 2008). These interactions are particularly strong for pharmaceuticals with multiple aromatic rings or extended conjugated systems, dominating for high-temperature biochars with extensive aromaticity (Tong et al., 2019).

Hydrogen bonding between polar functional groups in pharmaceuticals and oxygen-rich biochar surface sites contributes substantially. This is particularly true for polar compounds and low-temperature biochars retaining abundant surface functionalities (Yao et al., 2012). This mechanism operates across a wider pH range than electrostatic interactions (Ndoun et al., 2021).

Electrostatic interactions depend on solution pH and ionization state of both pharmaceutical and biochar surface. When the pH exceeds the pKa of the pharmaceutical, anionic pharmaceuticals such as sulfamethoxazole (pKa 1.6 and 5.7) become predominantly deprotonated and experience electrostatic repulsion from negatively charged biochar surfaces under alkaline conditions (Zheng et al., 2013). This pH-dependent behavior means that strong adsorption in the laboratory at pH 2 to 3 may not translate to comparable performance at neutral pH.

Hydrophobic partitioning contributes for non-polar compounds. Adsorption capacity correlates to octanol/water partition coefficient, with compounds having log Kow > 2.5 showing enhanced adsorption (Ndoun et al., 2021). Pore filling provides physical retention when pharmaceutical molecular dimensions are comparable to pore sizes (Yao et al., 2012).

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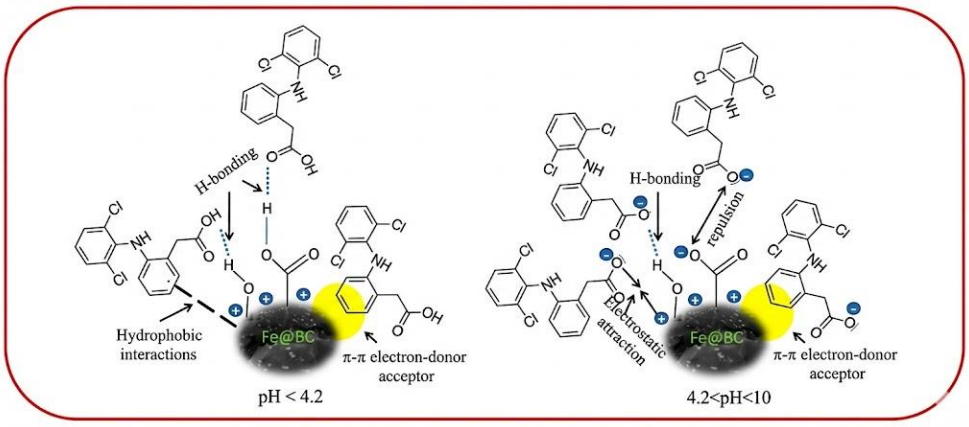

Figure 4. Adsorption mechanisms of pharmaceuticals on biochar at different pH conditions.

Source: Tam et al., 2020

3.3 Performance and Capacity Comparison

Reported adsorption capacities for individual pharmaceuticals on biochar vary widely, reflecting differences in biochar preparation, operational pH, and contact time. **Diclofenac**, a hydrophobic, ionizable pharmaceutical, shows reported maximum capacities ranging from 50 to 877 mg/g. This reflects optimization at very favorable conditions ($ZnCl_2$-activated biochar, pH 2 to 3) versus more realistic scenarios (Ndoun et al., 2021). Practical capacities at neutral pH on conventional biochar typically fall in the range of 50 to 200 mg/g.

Ibuprofen similarly shows wide variation from 10 to 570 mg/g depending on biochar type. Steam-activated materials generally outperform non-activated biochar (Tong et al., 2019). Real wastewater performance at neutral pH is substantially lower (10 to 100 mg/g) because ibuprofen deprotonates above pH 5 and experiences electrostatic repulsion (Tan et al., 2016).

Carbamazepine, a neutral pharmaceutical with minimal charge across operational pH ranges, achieves moderate capacities (5 to 90 mg/g) through π-π interactions and hydrogen bonding (Ndoun et al., 2021). Performance is relatively independent of solution pH, making it more suitable for direct application in neutral wastewater (Chen et al., 2008).

Sulfamethoxazole shows consistently low capacity (<40 mg/g at neutral pH) because its anionic character above pH 6 creates electrostatic repulsion (Zheng et al., 2013).

Table 3. Adsorption Capacity Comparison: Biochar vs. Activated Carbon (at neutral pH)

Pharmaceutical	Log Kow	pKa	Biochar (mg/g)	Activated Carbon (mg/g)	Biochar Advantage
Diclofenac	4.51	4.15	50 - 200	200 - 450	Lower cost for moderate removal
Ibuprofen	3.97	4.91	10 - 100	300 - 600	Biochar underperforms
Carbamazepine	2.45	13.9	5 - 50	80 - 200	Similar pH independence
Sulfamethoxazole	0.89	1.6/5.7	2 -20	40 - 150	Both perform poorly
Naproxen	3.18	4.15	30 - 150	150 - 350	Comparable performance

When biochar capacity is normalized to cost, biochar becomes economically superior to activated carbon for moderately hydrophobic pharmaceuticals (log Kow > 2.5), achieving 70 to 90% removal at a fraction of the media cost.

3.4 Real Wastewater Performance and Practical Constraints

Laboratory adsorption tests using pharmaceutical-spiked clean water systematically overestimate field performance because dissolved organic matter in real wastewater competes for adsorption sites (Tan et al., 2016). Pilot-scale studies treating actual secondary effluent commonly report pharmaceutical removal 30 to 70% lower than predictions from clean-water isotherms (Ndoun et al., 2021). Biochar contactors treating municipal effluent continuously reach saturation within 2 to 6 weeks of operation (Yao et al., 2012).

Regeneration addresses capacity exhaustion. Thermal regeneration at 400 to 600°C recovers 60 to 90% of original capacity over multiple cycles but requires dedicated equipment and energy investment. This is economically viable only at treatment scales exceeding approximately 1000 m^3/day (Tan et al., 2016). Chemical regeneration generates secondary waste requiring disposal. Many operators favor disposable biochar use, though this approach diminishes sustainability advantages (Yao et al., 2012).

Practical operation requires upstream pretreatment to prevent excessive suspended solids and biological growth from clogging the biochar bed (Tan et al., 2016). Integration downstream of conventional secondary treatment provides protection through removal of bulk suspended matter (Ndoun et al., 2021).

3.5 Strengths and Limitations

Biochar adsorption offers compelling advantages for cost and energy-constrained settings. Production from agricultural waste ($100 to 300 per ton) and operation requiring minimal energy (<0.1 kWh/m^3) make the technology accessible in rural and off-grid contexts (Yao et al., 2012). Operator skill requirements are low. Primary tasks involve pump operation, periodic media replacement, and backwash frequency adjustment (Tan et al., 2016). Biochar demonstrates superior cost-effectiveness for hydrophobic pharmaceuticals (log Kow > 2.5), achieving 70 to 95% removal for NSAIDs at costs substantially lower than competing technologies (Tong et al., 2019).

Limitations are material. Performance for polar and anionic pharmaceuticals is weak without pH adjustment (Ndoun et al., 2021). Biochar quality is variable across batches, requiring characterization testing before deployment (Tan et al., 2016). Saturation and spent biochar disposal present challenges if regeneration is not economically justified (Yao et al., 2012).

4. MEMBRANE FILTRATION

4.1 Membrane Technology Spectrum

Pressure-driven membrane processes separate pharmaceuticals based on size exclusion and electrostatic interactions across a spectrum of pore sizes. Microfiltration (MF, 0.1 to 10 μm pore size) and ultrafiltration (UF, 1 to 100 nm) primarily remove particulates, colloids, and microorganisms, achieving only indirect pharmaceutical removal through association with colloidal matter (Kimura et al., 2005). Their principal role in pharmaceutical wastewater treatment is pretreatment protection against fouling of downstream NF or RO (Bellona et al., 2004).

Nanofiltration (NF, 0.5 to 10 nm pores, 150 to 300 Da molecular weight cutoffs) provides pharmaceutical-specific rejection through size exclusion and electrostatic repulsion (Bellona et al., 2004). Most NF membranes carry net negative charge at neutral pH, creating electrostatic barriers to anionic pharmaceuticals while showing weaker rejection of neutral and cationic species (Kimura et al., 2005).

Reverse osmosis (RO, <0.1 nm effective pore size, <100 Da cutoff) achieves near-universal rejection of all dissolved solutes through a solution-diffusion transport mechanism (Luo et al., 2014). RO provides highly non-selective rejection across pharmaceutical classes but at substantially higher operating pressure.

Energy demand scales with operating pressure. NF typically requires 0.5 to 2.0 kWh/m^3 at 75% water recovery, while RO demands 1.5 to 6.0 kWh/m^3 under equivalent conditions (Kimura et al., 2005).

4.2 Pharmaceutical Rejection Performance

Nanofiltration rejection of pharmaceuticals varies with both membrane characteristics and compound properties. Anionic pharmaceuticals at neutral pH (such as diclofenac, deprotonated above pH 4) experience strong electrostatic repulsion from negatively charged NF membranes, typically achieving 75 to 95% rejection on NF270 and >95% rejection on NF90 (Bellona

et al., 2004). Neutral pharmaceuticals such as carbamazepine (236 Da) show size-dependent rejection: approximately 40 to 70% on NF270 and 75 to 90% on NF90 (Kimura et al., 2005). Compounds smaller than approximately 180 Da generally show lower rejection.

Operating conditions, particularly transmembrane pressure and feed pH, further modulate NF rejection. Higher pressure generally increases rejection by reducing solute back-diffusion, while pH alters both pharmaceutical speciation and membrane surface charge, strengthening or weakening electrostatic exclusion for ionizable compounds (Figure 5).

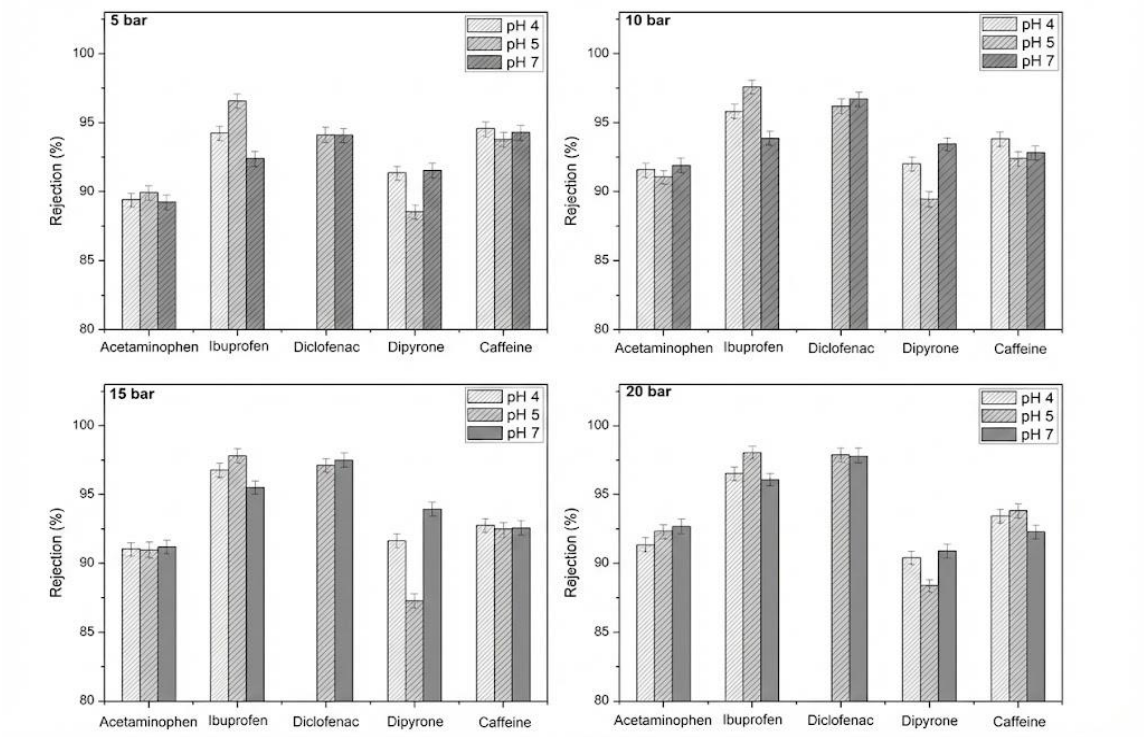

Figure 5. Rejection of selected pharmaceuticals (acetaminophen, ibuprofen, diclofenac, dipyrone and caffeine) by a nanofiltration membrane as a function of transmembrane pressure (5, 10, 15 and 20 bar) and feed pH (4, 5 and 7)

Reverse osmosis provides uniform and high rejection across all pharmaceutical classes. Pilot-scale RO treatment of municipal secondary effluent demonstrated >98% removal for carbamazepine, sulfamethoxazole, diclofenac, and ibuprofen at typical flux rates of 15 to 25 L/m²·h and water recovery of 75 to 85% (Kimura et al., 2005). The consistency of RO rejection across diverse pharmaceutical structures eliminates the need for compound-specific optimization (Bellona et al., 2004).

Table 4. Membrane Pharmaceutical Rejection by Type and Compound

Pharmaceutical	MW (Da)	Charge at pH 7	NF90 Rejection	NF270 Rejection	RO Rejection	Mechanism
Carbamazepine	236	Neutral	75 - 90%	40 - 70%	>98%	Size exclusion
Diclofenac	296	Anionic	>95%	75 - 88%	>99%	Size + electrostatic
Ibuprofen	206	Anionic	85 - 95%	70 - 85%	>98%	Size + electrostatic
Sulfamethoxazole	253	Mixed	80 - 90%	50 - 75%	>95%	Size exclusion
Atenolol	266	Cationic	85 - 92%	60 - 80%	>97%	Size + weak electrostatic

4.3 Fouling and Operational Challenges

Membrane fouling is the primary operational challenge in pharmaceutical wastewater treatment. Organic matter in municipal secondary effluent, typically 5 to 30 mg/L of dissolved organic carbon, deposits on and within membrane pores. This organic matter comprises primarily humic substances and soluble microbial products (Kimura et al., 2005). Flux decline of 20 to 50% commonly occurs within 24 to 72 hours of continuous operation without effective upstream pretreatment (Bellona et al., 2004).

Inorganic fouling results from precipitation of calcium, magnesium, and silica salts when concentration polarization at the membrane surface raises local saturation indices (Luo et al., 2014). Biofouling develops as bacteria colonize membrane surfaces and produce biofilms stabilized by extracellular polymeric substances (Kimura et al., 2005).

Membrane lifespan in pharmaceutical wastewater applications averages 3 to 5 years of continuous operation, compared to 5 to 10 years in drinking water applications (Bellona et al., 2004). Chemical cleaning protocols employing sodium hypochlorite (2000 ppm), citric acid (2000 ppm), and methanol recover 40 to 90% of flux but gradually degrade polymer structure (Kimura et al., 2005).

4.4 Energy and Cost Considerations

Energy demand for NF treating municipal effluent at moderate recovery (75%) ranges from 0.5 to 2.0 kWh/m³ (Bellona et al., 2004). RO requires substantially higher energy: 1.5 to 4.0 kWh/m³ for standard operating conditions, extending to 3 to 6 kWh/m³ when treating heavily fouled influent or operating at lower recovery ratios (Luo et al., 2014). These energy demands exceed ozonation (0.05 to 0.30 kWh/m³) and biochar (<0.1 kWh/m³) by orders of magnitude.

Capital costs for municipal-scale membrane systems (treating 1,000 to 10,000 m³/day) range from $500 to 1500 per m³/day capacity (Luo et al., 2014). Operating costs span $0.20 to 0.80/m³, distributed among energy (40 to 60% of OPEX), membrane replacement (20 to 35%), chemical cleaning (10 to 20%), and labor (5 to 15%) (Bellona et al., 2004). These costs substantially exceed conventional treatment ($0.05 to 0.15/m³) and biochar adsorption ($0.05 to 0.20/m³).

Concentrate management adds significant cost. NF and RO generate concentrated brine (15 to 25% of influent volume at 3 to 8× feed concentration) (Luo et al., 2014). Inland facilities without saline outfalls face disposal costs of $1 to 5/m³, potentially doubling total treatment cost (Kimura et al., 2005).

4.5 Integration in Treatment Trains

Membrane bioreactors integrate biological treatment with submerged MF or UF, combining biodegradation with membrane separation (Luo et al., 2014). MBR pharmaceutical removal ranges from 30% to >99% depending on compound biodegradability. Readily degradable ibuprofen achieves 90 to 99% removal while recalcitrant carbamazepine remains <30% (Kimura et al., 2005). MBRs provide robust pretreatment for downstream RO by eliminating suspended solids (Bellona et al., 2004).

MBR-RO systems create multi-barrier configurations for potable reuse, achieving >99.5% pharmaceutical removal across all compounds, complete pathogen elimination (>6 log reduction), and dissolved organic carbon reduction below 10 mg/L (Luo et al., 2014). Superior pretreatment from MBRs extends RO membrane intervals and lifespan by 30 to 50% (Kimura et al., 2005).

Hybrid AOP-membrane configurations combine oxidative pretreatment with membrane polishing. Ozonation upstream of NF or RO oxidizes recalcitrant pharmaceuticals and reduces dissolved organic matter causing fouling, while downstream membranes eliminate transformation products and provide redundant pharmaceutical barriers (Luo et al., 2014). Pilot studies document 15 to 20% improvements in permeate pharmaceutical concentration when ozonation precedes NF (Hollender et al., 2009).

5. CROSS-TECHNOLOGY PERFORMANCE COMPARISON

5.1 Harmonized Performance Metrics

Meaningful technology comparison requires standardized metrics. Removal efficiency alone is insufficient because the lowest-cost technology with adequate performance often represents better value than the highest-performance option if regulatory targets are modest.

Energy consumption varies by multiple orders of magnitude. Biochar requires <0.1 kWh/m³, ozonation 0.05 to 0.30 kWh/m³ (Reungoat et al., 2012), photo-Fenton and NF 0.5 to 2.0 kWh/m³ (Antonopoulou et al., 2021), and RO 1.5 to 6.0 kWh/m³ (Luo et al., 2014). This range suggests technology selection should strongly consider electricity availability and cost in energy-constrained regions.

Treatment costs show corresponding variation. Biochar systems cost $0.05 to 0.20/m³ (Yao et al., 2012), ozonation $0.10 to 0.30/m³ (Reungoat et al., 2012), AOPs $0.30 to 1.00/m³ (Antonopoulou et al., 2021), and membrane systems $0.20 to 0.80/m³ (Luo et al., 2014). These ranges reflect the cost/performance continuum. Lowest-cost biochar achieves moderate removal (70 to 90%) suitable for discharge, while highest-cost RO justifies premium through consistent >95% removal across all pharmaceutical classes (Kimura et al., 2005).

Capital costs range from $200 to 600 per m³/day for biochar through $800 to 1500 per m³/day for RO, influencing affordability in capital-constrained municipalities.

Technology readiness varies. Ozonation and RO operate at full scale globally with decades of experience (TRL 9), biochar remains mostly pilot-scale (TRL 6 to 7), while photo-Fenton and photocatalysis remain largely demonstration-scale (TRL 5 to 6).

5.2 Comprehensive Technology Comparison

Table 5. Cross-Technology Performance Comparison

Criterion	Ozonation	Photo-Fenton	Biochar	NF	RO
Removal Efficiency (%)	70 - 85	>95	70 - 90*	50 - 95	>95
Energy (kWh/m³)	0.05 - 0.30	0.50 - 2.00	<0.10	0.50 - 2.00	1.5 - 6.0
Cost ($/m³)	0.10 - 0.30	0.50 - 1.00	0.05 - 0.20	0.30 - 0.60	0.40 - 0.80
CAPEX ($/m³/day)	400 - 800	600 - 1200	200 - 600	500 - 1200	800 - 1500
Operational Complexity	Medium	High	Low	Medium–High	High
TRL	9	5 - 6	6 - 7	8 - 9	9
TP Risk	Medium - High	Low	None	None	None

*For hydrophobic pharmaceuticals; <40% for anionic compounds at neutral pH

5.3 Compound-Specific Performance and Technology Matching

Removal efficiency for specific pharmaceuticals varies substantially across technologies, supporting compound-matched technology selection. Carbamazepine, a recalcitrant neutral compound showing <20% conventional treatment removal, achieves 80 to 95% ozonation removal (Reungoat et al., 2012), >95% photo-Fenton (Oller et al., 2011), 40 to 80% biochar (variable) (Ndoun et al., 2021), 50 to 80% NF (Bellona et al., 2004), and >98% RO (Kimura et al., 2005). When carbamazepine is a regulatory driver, photo-Fenton or RO are the only reliably compliant options.

Ibuprofen, readily biodegradable, shows excellent removal across all advanced technologies: >85% ozonation (Reungoat et al., 2012), >95% Fenton (Antonopoulou et al., 2021), 70 to 95% biochar (Tong et al., 2019), >85% NF (Bellona et al., 2004), >98% RO (Kimura et al., 2005). Ibuprofen removal is rarely limiting. Cost and energy typically dominate technology selection.

Hydrophobic NSAIDs (diclofenac, naproxen; log Kow > 3) show enhanced biochar adsorption (>80% removal), making biochar cost-effective first choice for this pharmaceutical class (Ndoun et al., 2021).

Anionic, hydrophilic pharmaceuticals (sulfamethoxazole; log Kow <1) show weak biochar performance (<40% at neutral pH) (Zheng et al., 2013) but strong NF and RO rejection (>80%), making membranes obligatory (Bellona et al., 2004).

6. HYBRID AND INTEGRATED SYSTEMS

6.1 Common Hybrid Configurations

Hybrid systems combine two or more technologies to leverage complementary mechanisms. Ozone + Biochar/GAC pairs oxidative pretreatment with adsorptive polishing. Ozonation oxidizes recalcitrant pharmaceuticals and bulk organic matter, while downstream biochar removes transformation products and residual pharmaceuticals (Reungoat et al., 2012).

AOP + Biological Treatment exploits oxidative enhancement of biodegradability. Partial oxidation by ozone, Fenton, or UV/H_2O_2 converts recalcitrant pharmaceuticals into biodegradable intermediates. Downstream biological reactors then mineralize these intermediates (Oller et al., 2011). This configuration reduces AOP chemical demand by 30 to 50% (Antonopoulou et al., 2021).

MBR-NF and MBR-RO integrate biological removal with membrane barriers. MBRs remove biodegradable compounds and provide superior pretreatment while NF or RO addresses recalcitrant compounds (Luo et al., 2014).

NF/RO + Upstream AOP or Biochar reduces membrane fouling and provides redundant pharmaceutical barriers. Upstream treatment reduces organic matter load (Bellona et al., 2004).

6.2 Performance Improvements and Synergistic Effects

Hybrid systems consistently outperform standalone technologies. Ozone + GAC achieves 85 to 95% total pharmaceutical removal compared to 60 to 80% for ozonation alone and 70 to 85% for GAC alone (Reungoat et al., 2012). The benefit stems from GAC adsorption of ozone transformation products. Ozonation alone generates TPs but GAC removes these byproducts, reducing ecotoxicological risk.

MBR-RO systems achieve >95% pharmaceutical removal across all compounds with complete pathogen elimination, meeting potable reuse standards. These systems also reduce fouling and extend RO membrane lifespan by 30 to 50% (Luo et al., 2014).

Photo-Fenton + Biological Post-treatment achieves >98% pharmaceutical mineralization at lower oxidant doses than photo-Fenton alone, reducing chemical costs by 30 to 50% (Oller et al., 2011).

Field-scale validation confirms these benefits. A Swiss WWTP upgraded with post-ozonation followed by sand filtration and granular activated carbon achieved >95% removal for 12 monitored pharmaceuticals over 2-year continuous operation (Hollender et al., 2009). An Australian MBR-RO reclamation facility demonstrates >99% pharmaceutical removal with stable performance across seasonal variations.

6.3 Implementation Trade-offs

Hybrid systems add capital and operational complexity. Ozone + GAC requires separate contactors and ozone generation equipment, increasing capital costs 40 to 70% versus standalone ozonation. However, operating costs increase only 20 to 30% due to reduced ozone demand (Reungoat et al., 2012). MBR-RO carries the highest capital costs ($1000 to 2500 per m^3/day), justified primarily for potable reuse applications where treated water value offsets desalination costs in water-scarce regions (Luo et al., 2014).

Hybridization is justified when stringent reuse standards require >95% removal across all compounds, hospital wastewater creates complex pharmaceutical mixtures, transformation product toxicity must be minimized, or sustainability requires resource recovery. For discharge-only applications with moderate limits (70 to 85% removal), single technologies often suffice at substantially lower cost.

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7. DECISION FRAMEWORK FOR TECHNOLOGY SELECTION

7.1 Key Decision Criteria

Successful technology selection balances multiple competing criteria. Treatment objective distinguishes discharge compliance (typically 60 to 80% removal acceptable) from non-potable reuse (>90% removal) and potable reuse (>95% removal with multi-barrier verification). Regulatory limits and target pharmaceuticals determine required removal. Figure 6 summarizes these criteria in a treatment objective-based decision tree for practical technology selection.

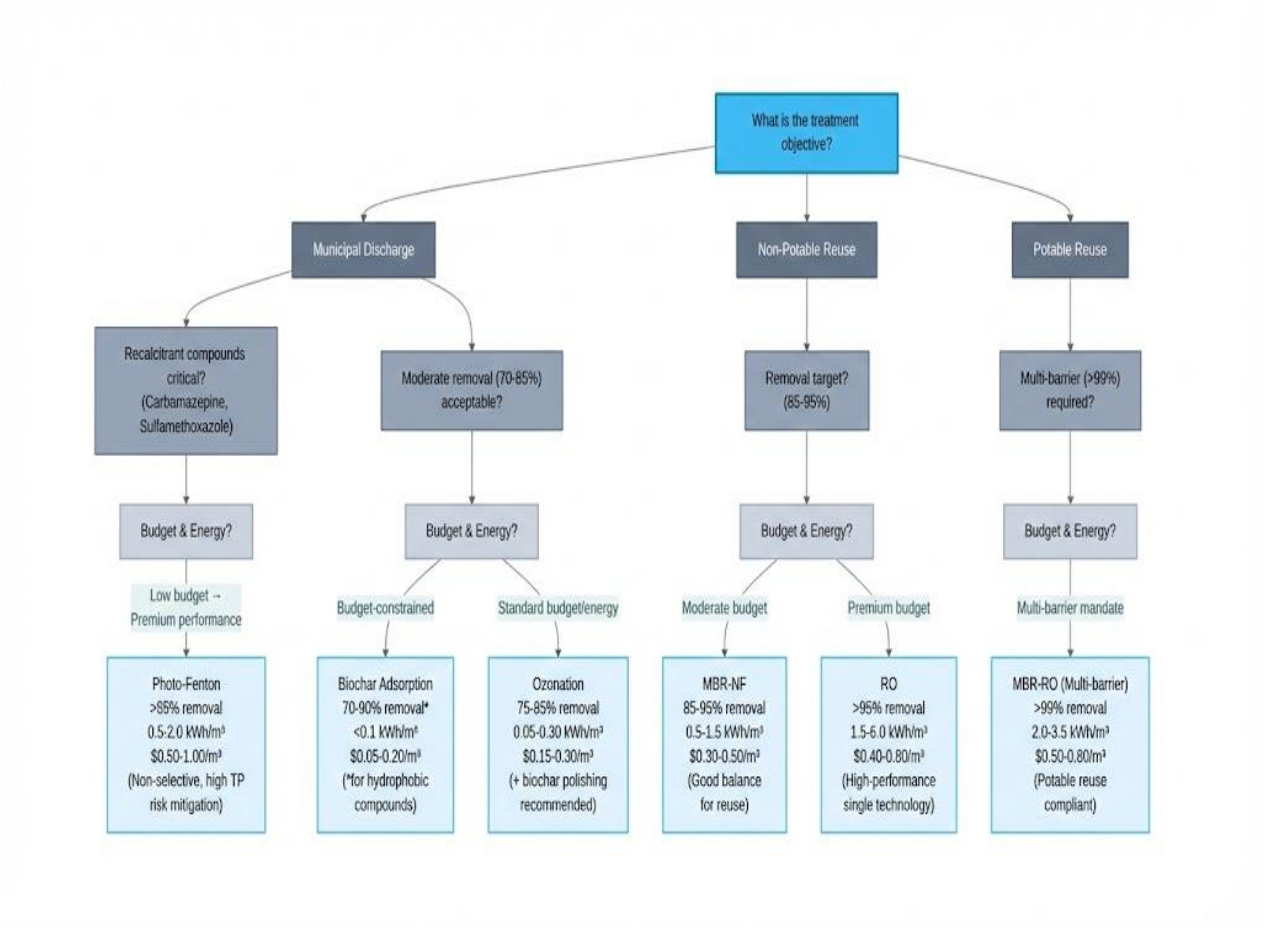

Figure 6. Decision tree for technology selection for pharmaceutical removal from municipal wastewater.

Budget and energy availability strongly constrain options. Capital-limited municipalities may be forced toward biochar ($200 to 600/m³/day) over RO ($800 to 1500/m³/day). Energy-constrained settings must favor biochar (<0.1 kWh/m³) and ozonation (0.05 to 0.30 kWh/m³) over RO (1.5 to 6.0 kWh/m³).

Existing infrastructure influences retrofit decisions. Existing UV systems can add H_2O_2 for AOP with modest investment. Local context including workforce skills, chemical supply chains, and waste disposal options determines operational feasibility. Resource-limited settings favor simpler biochar systems. Regions with established ozone use in drinking water can replicate expertise.

7.2 Multi-Criteria Decision Analysis Methodology

The qualitative pathways in Figure 6 are formalized here using a multi-criteria decision analysis. Structured decision frameworks weight criteria reflecting local priorities: removal efficiency 20 to 40% when regulatory limits are stringent, cost 20 to 35% reflecting budget constraints, energy 10 to 20%, operational complexity 10 to 15%, technology readiness 5 to 15%, and transformation product risk 5 to 10%. Each technology scores 1 to 5 for each criterion. Weighted scores yield overall ranking for specific scenarios.

7.3 Scenario-Based Recommendations

Table 6. Technology Recommendations by Operational Context

Application Scenario	Primary Recommendation	Removal Target	Typical Cost ($/m³)	Justification
Municipal WWTP discharge upgrade	Ozonation	70 - 80%	0.15 - 0.30	Proven, cost-effective
Tertiary treatment (high removal)	RO or Photo-Fenton	>95%	0.40 - 1.00	Non-selective, reliable
Hospital pharmaceutical wastewater	Photo-Fenton	>95%	0.50 - 1.00	Non-selective; complex mixtures
Off-grid/rural facility	Biochar + solar photocatalysis	70 - 85%	0.05 - 0.15	Minimal energy
Non-potable reuse (irrigation)	MBR-NF or Ozone + Biochar	85 - 95%	0.25 - 0.50	Moderate removal; cost balance
Potable reuse	MBR-RO	>99%	0.50 - 0.80	Multi-barrier; compliance

Municipal wastewater treatment plant upgrades for discharge compliance should consider ozonation as the primary option. Ozonation offers proven performance (75 to 85% removal) (Reungoat et al., 2012), established operational protocols, and moderate costs (Luo et al., 2014). Where transformation product concerns exist or >85% removal is required, ozone + biochar polishing provides a robust solution.

New plants designed for non-potable reuse should evaluate MBR-NF combinations, achieving 85 to 95% removal with energy requirements 40 to 50% lower than RO (Luo et al., 2014). Biochar polishing after MBR provides lower-cost alternatives where NF-level performance suffices.

Hospital and high-concentration pharmaceutical wastewaters should select photo-Fenton or electrochemical oxidation, providing non-selective removal suitable for complex pharmaceutical mixtures (Antonopoulou et al., 2021).

Resource-constrained municipalities should prioritize biochar adsorption using locally-sourced agricultural waste, achieving 70 to 80% removal for hydrophobic pharmaceuticals at minimal capital cost and zero grid electricity requirement (Yao et al., 2012).

8. RESEARCH GAPS AND FUTURE DIRECTIONS

8.1 Identified Knowledge Gaps

AOP transformation products have been extensively characterized chemically but lack full-scale ecotoxicological validation. Most pilot studies measure parent pharmaceutical removal without comprehensive TP identification or bioassay screening. Long-term environmental monitoring downstream of AOP-equipped facilities remains a critical gap (Escher & Fenner, 2011).

Biochar adsorption literature emphasizes controlled laboratory conditions but inadequately addresses dissolved organic matter competition in real wastewater. Quantitative models predicting pharmaceutical removal reduction in actual effluent based on NOM composition remain unavailable (Tan et al., 2016).

Membrane fouling research focuses on bulk organic matter without characterizing pharmaceutical-specific interactions or addressing concentrate treatment strategies for pharmaceutical-laden brines (Bellona et al., 2004).

Hybrid system mechanisms remain poorly understood. Why do certain combinations outperform individual process predictions? How do transformation products from AOPs interact with downstream biological or adsorptive processes? These questions require detailed investigation (Oller et al., 2011).

8.2 Emerging Technologies

Biochar-photocatalyst composites combining adsorption with in situ photocatalytic degradation show laboratory promise. These materials enable pharmaceutical concentration followed by light-activated oxidation that could enable regeneration without thermal treatment (Tan et al., 2016).

Integrated AOP-membrane bioreactors incorporate oxidative treatment within biological reactors, combining degradation benefits with separation in compact footprints. Challenges include preventing oxidant-induced biomass inactivation (Oller et al., 2011).

Low-energy membranes using forward osmosis, electrodialysis, or biomimetic aquaporin membranes promise 50 to 70% energy reduction versus RO (Kimura et al., 2005). Pharmaceutical rejection performance of novel materials requires characterization.

Digital monitoring and optimization employing real-time pharmaceutical sensors, machine learning for oxidant dosing, and predictive fouling models could substantially improve operational efficiency (Luo et al., 2014).

Table 7. Research Gap Summary with Implementation Priorities

Gap	Current Understanding	Research Need	Priority	Implementation Method
TP Ecotoxicity	Chemical characterization available	Full-scale bioassay validation	High	Environmental monitoring + effect-based testing
Biochar Economics	Regeneration possible but costly	Cost-benefit analysis at scale	High	Pilot demonstrations + economic modeling
Membrane Fouling	NOM drives decline	Pharmaceutical-specific mechanisms	High	Controlled fouling studies
NOM Competition	30 - 70% capacity loss observed	Predictive models for real water	Medium	Competitive isotherm studies
Hybrid Synergy	10 - 20% improvement observed	Mechanistic understanding	Medium	Molecular-level analysis

9. CONCLUSIONS

9.1 Synthesis of Findings

No single "best" technology exists. Context determines optimal choice through deliberate analysis weighing technical performance, cost, energy, and operational capacity. RO and photo-Fenton achieve highest removal (>95%) but demand substantial energy (1.5 to 6.0 and 0.5 to 2.0 kWh/m³) and cost ($0.40 to 1.00/m³). These technologies are justifiable for potable reuse and stringent applications but economically irrational for moderate-performance requirements.

Biochar adsorption provides the most cost-effective solution ($0.05 to 0.20/m³) for moderate removal (70 to 90% for hydrophobic pharmaceuticals). It is suited for resource-limited settings and discharge-only applications. Performance is compound-dependent and degrades with dissolved organic matter competition.

Ozonation offers proven middle-ground performance (75 to 85% removal) at moderate cost ($0.15 to 0.30/m³) and energy (0.05 to 0.30 kWh/m³) with established global operational experience. Transformation product formation requires downstream polishing or biological post-treatment.

Hybrid systems consistently outperform standalone technologies by 10 to 20 percentage points. Added complexity justifies investment when stringent removal, reuse applications, or sustainability requirements demand robust multi-barrier approaches.

9.2 Practical Recommendations

Municipal WWTP discharge upgrades should select ozonation or biochar based on regulatory removal targets and local energy/cost constraints (Reungoat et al., 2012). Non-potable reuse applications should employ MBR-NF or ozone + biochar combinations achieving 85 to 95% removal at acceptable cost (Luo et al., 2014). Potable reuse demands MBR-RO or equivalent multi-barrier configurations (Kimura et al., 2005). Hospital wastewater requires photo-Fenton or equivalent non-selective oxidation (Antonopoulou et al., 2021). Resource-constrained rural settings should implement biochar from local waste feedstocks (Yao et al., 2012).

Regulatory frameworks should shift from simple concentration-based pharmaceutical limits toward risk-based approaches incorporating transformation products and effect-based bioassays (Escher & Fenner, 2011).

9.3 Final Perspective

Future pharmaceutical removal will employ technology combinations tailored to specific contexts rather than universal solutions. Water-scarce regions will increasingly adopt MBR-RO for integrated treatment and reuse offsetting supply costs. Energy-constrained developing nations will implement biochar from local biomass, creating livelihoods around waste valorization. Advanced economies with stringent regulations will deploy ozone-biological-membrane treatment trains. Climate change and population growth will elevate pharmaceutical removal from optional advanced treatment to integral water security strategy.

Research priorities should focus on reducing energy intensity of high-performance technologies, validating transformation product risk at full scale, and developing decision support tools for technology transfer across diverse global contexts.

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