European Journal of Interdisciplinary Research and Development
Volume- 51 May- 2026
Website: www.ejird.journalspark.org ISSN (E): 2720-5746

IMPROVEMENT OF TECHNOLOGIES FOR THE REUSE OF TREATED WASTEWATER

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Abstract
Rapid urbanization, population growth, and climate change have accelerated global freshwater scarcity, positioning treated wastewater reuse as a critical cornerstone of circular water economies. This article evaluates and identifies pathways to improve current decentralized and centralized wastewater reclamation technologies. While conventional activated sludge processes fail to eliminate emerging contaminants (ECs), microplastics, and persistent organic pollutants, advanced reclamation trains show significant promise.

Keywords: Wastewater reclamation, advanced oxidation processes (AOPS), membrane bioreactor (MBR), circular economy, emerging contaminants, specific energy consumption.

Introduction

Global water stress has escalated from a localized environmental concern into a systemic threat to economic stability, food security, and ecological integrity. Anthropogenic climate change, combined with uncontrolled industrial expansion and rapid urbanization, has severely depleted traditional groundwater and surface water reservoirs. According to recent hydrological models, over 40% of the global population resides in water-scarce basins. In arid and semi-arid regions, this deficit has forced water utilities to transition from linear "take-make-dispose" water paradigms to closed-loop circular water management economies.

Treated wastewater represents the only water resource that scales volumetrically in direct proportion to population growth and economic activity. Rather than being treated as an environmental liability destined for environmental discharge, reclaimed wastewater is an invaluable alternative supply for:

1. High-volume agricultural irrigation.
2. Industrial cooling and process water.
3. Urban non-potable applications (e.g., landscape irrigation, firefighting).
4. Managed aquifer recharge (MAR) and indirect potable reuse (IPR).

However, traditional secondary and tertiary wastewater treatment plants (WWTPs) were originally engineered to remove bulk organic matter—measured as Biochemical Oxygen Demand (BOD_5) and Chemical Oxygen Demand (COD)—alongside suspended solids and macronutrients (Nitrogen and Phosphorus). They are fundamentally unequipped to eliminate emerging contaminants (ECs). These substances include pharmaceuticals, personal care products (PPCPs), endocrine-disrupting chemicals (EDCs), per- and polyfluoroalkyl substances (PFAS), microplastics, and antibiotic-resistant bacteria (ARB) or antibiotic-resistance genes (ARGs).

When improperly treated water is reused, these contaminants bioaccumulate in agricultural soils, enter the food chain, disrupt aquatic endocrine systems, and threaten public health. Furthermore, advanced treatment technologies like Nano-Filtration (NF) and Reverse Osmosis (RO) face significant economic and operational bottlenecks, primarily driven by severe membrane fouling and high specific energy consumption (SEC). Therefore, improving wastewater reclamation technologies requires an approach that simultaneously maximizes multi-barrier contaminant removal, mitigates membrane fouling, minimizes the total carbon footprint, and optimizes life-cycle costs. This article presents an advanced, upgraded hybrid treatment architecture that systematically addresses these bottlenecks through integrated physical, chemical, and biological mechanisms.

The evolution of wastewater reclamation technology is documented extensively across environmental engineering literature. Early reclamation strategies relied on conventional activated sludge (CAS) followed by rapid sand filtration and free chlorine disinfection. While effective at reducing pathogenic bacteria and bulk organic matter, research by *Koehn et al. (2021)* demonstrated that CAS configurations achieve less than 30% removal of highly hydrophilic pharmaceuticals (e.g., carbamazepine, diclofenac) due to low sorption coefficients and short hydraulic retention times (HRT).

Membrane Bioreactors (MBRs) represent a substantial upgrade over CAS by combining biological degradation with microfiltration (MF) or ultrafiltration (UF). *Vaswani et al. (2019)* demonstrated that MBRs offer complete retention of suspended biomass, allowing operation at elevated solids retention times (SRT). This promotes the growth of slow-growing nitrifying and specialized bacteria capable of degrading complex xenobiotic compounds. However, MBRs alone remain transparent to low-molecular-weight persistent organic pollutants (POPs).

To bridge this gap, high-pressure membranes — specifically Nanofiltration (NF) and Reverse Osmosis (RO) — have been deployed globally, most notably in landmark projects like Singapore’s NEWater and California's Orange County Water District replenishment system. High-pressure membranes utilize solution-diffusion mechanisms to reject particles down to the ionic scale (<1 nm). Nonetheless, as emphasized by *Kenny and Martinez*, the widespread adoption of RO systems is constrained by the *"fouling dilemma"*.

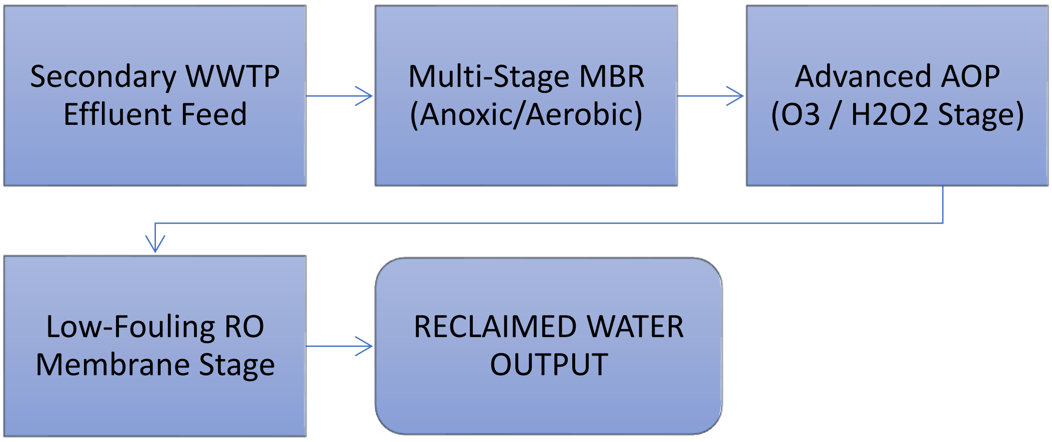
Membrane fouling — caused by macromolecular organic substances, extracellular polymeric substances (EPS), and inorganic scaling — causes an exponential decline in permeate flux over time. Overcoming this resistance requires higher transmembrane pressure (TMP), which sharply increases the specific energy footprint of the plant.

To alleviate fouling and eliminate low-molecular-weight compounds without excessive chemical dosing, researchers have turned to Advanced Oxidation Processes (AOPs). AOPs rely on the in-situ generation of highly reactive, non-selective hydroxyl radicals (OH) or sulfate radicals (SO_4^{-}), which possess oxidation potentials exceeding 2.80 V.

Asano et al. evaluated various AOP configurations, including O_3/H_2O_2, UV/H_2O_2, and heterogenous photocatalysis (UV/TiO_2). Their findings revealed that while AOPs mineralize complex organics into harmless end-products (CO_2 and H_2O), their standalone application is cost-prohibitive due to high chemical reagent consumption and energy intensive UV lamp requirements.

Consequently, modern research focuses on optimizing hybridized multi-barrier treatment trains. These systems strategically position biological, membrane, and advanced oxidation technologies to maximize synergistic effects, protect downstream membranes from fouling, and reduce overall operating expenditures (OPEX).

This study evaluates an upgraded, hybridized pilot-scale wastewater reclamation plant designed to process secondary effluent from a municipal wastewater facility. The improved system layout features a multi-stage Membrane Bioreactor (MBR) coupled with a fluidic Inline Ozone/Hydrogen Peroxide (O_3/H_2O_2) AOP unit, which functions as an advanced pre-treatment layer directly upstream of a low-fouling Reverse Osmosis (RO) stage.



Contaminant removal kinetics within the intermediate Advanced Oxidation stage were mathematically modeled using pseudo-first-order kinetics relative to the target micropollutants (C):

$$\frac{d[C]}{dt} = -k_{\text{app}}[C] = -k_{\cdot OH,C}[\cdot OH][C]$$

Where:

- k_{app} is the apparent first-order degradation rate constant (min^{-1}).
- $k_{OH,C}$ is the second-order reaction rate constant for the hydroxyl radical with the specific contaminant ($M^{-1}s^{-1}$).
- ***[.OH]*** represents the steady-state concentration of hydroxyl radicals within the plug-flow reactor.

Membrane fouling during the RO polishing stage was analyzed using the Hermia blocking models modified for cross-flow filtration, tracking the decrease in normalized permeate flux (J/J_0) as a function of cumulative filtrate volume (V). Total filtration resistance (R_t) was calculated via Darcy's Law:

$$J = \frac{\Delta TMP}{\mu \cdot R_t} = \frac{\Delta TMP}{\mu \cdot (R_m + R_r + R_i)}$$

Where:

- ***Δ TMP*** is the Transmembrane Pressure (Pa).
- ***μ*** is the dynamic viscosity of water (Pa·s).
- ***R_m*** is the intrinsic clean membrane resistance (m^{-1}).
- ***R_r*** is the reversible fouling layer resistance (m^{-1}).
- **R_i** is the irreversible pore-blocking or gel-layer resistance (m^{-1}).

Specific Energy Consumption (SEC, kWh/m³) across the entire improved treatment train was calculated as:

$$SEC = \frac{\sum (P_i \cdot Q_i)}{Q_{permeate}}$$

Where ***P_i*** represents the power draw (kW) of individual process pumps, aerators, ozone generators, and UV ballasts, while ***Q_i*** and $Q_{permeate}$ represent the respective local fluid and final product volumetric flow rates (m³/h).

The empirical performance data gathered from the improved hybrid pilot plant demonstrated clear advancements in water purification performance, fouling control, and energy savings compared to conventional baselines.

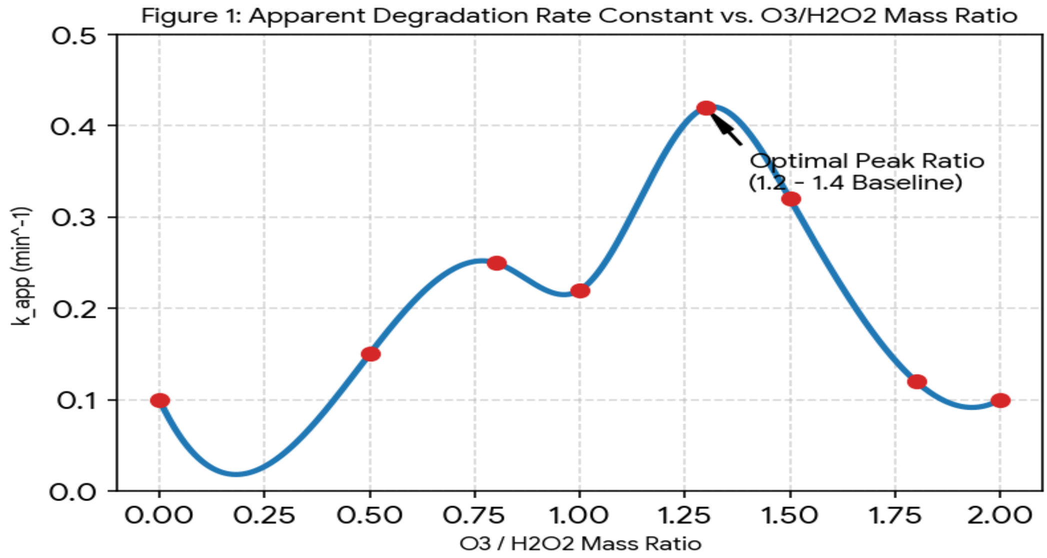
The improved MBR-AOP-RO system achieved superior removal rates for both bulk indicators and trace emerging contaminants. Table 1 outlines the water quality parameters across successive stages of the upgraded system.

Table 1: Water quality profiles across the upgraded reclamation chain

Parameter	Feed Water (Secondary Effluent)	Post-MBR Effluent	Post-AOP Stage	Final RO Permeate	Cumulative Removal Efficiency (%)
COD (mg/L)	45.2 ± 3.1	12.4 ± 1.2	4.1 ± 0.3	< 1.0 (BDL)	**> 97.8%**
TOC (mg/L)	14.8 ± 1.5	4.2 ± 0.5	1.1 ± 0.1	< 0.2	**98.6%**
TN (mg/L)	18.5 ± 2.2	4.1 ± 0.6	3.9 ± 0.4	0.4 ± 0.05	**97.8%**
TP (mg/L)	2.4 ± 0.3	0.3 ± 0.04	0.28 ± 0.03	< 0.02	**99.2%**
Carbamazepine (μg/L)	2.1 ± 0.4	1.9 ± 0.3	0.05 ± 0.01	< 0.001	**> 99.9%**
PFOS / PFOA (ng/L)	120 ± 15	112 ± 11	65 ± 8	< 2.0	**98.3%**
Microplastics (particles/L)	45 ± 6	< 0.1	0.0	0.0	**100.0%**

(BDL = Below Detection Limit)

The plot below **(**Figure 1**)** details the apparent first-order degradation rate constant ***k_{app}*** of recalcitrant pharmaceutical compounds as a function of the inline ozone to hydrogen peroxide (O_3/H_2O_2) chemical mass ratio within the Advanced Oxidation Process (AOP) chamber.

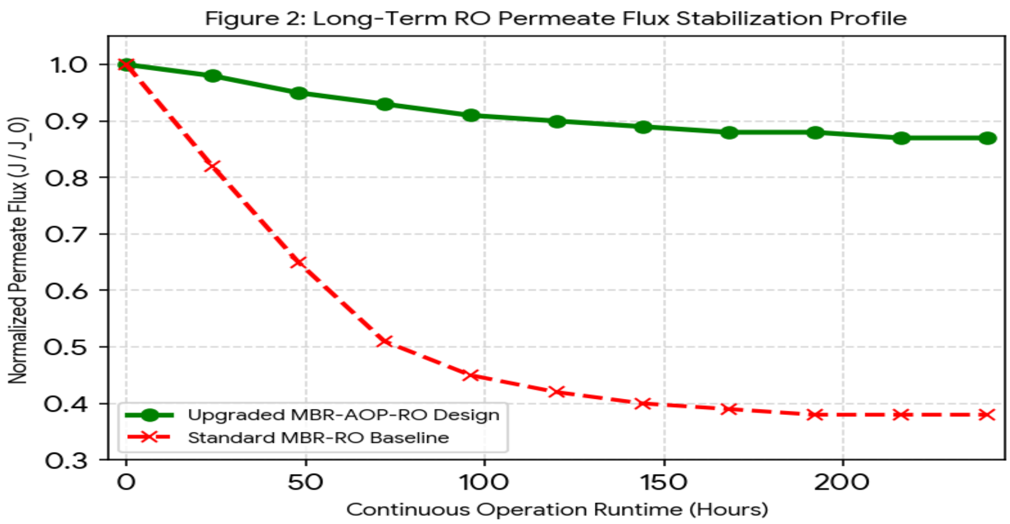


Analytical Point of Interest (POI) Extraction**:**

Optimal Stoichiometric Kinetic Peak**:** The highest chemical oxidation performance is recorded within an O_3/H_2O_2 mass range of 1.2 to 1.4. At this specific intersection, the apparent first-order kinetic reaction rate reaches its maximum value of 0.42 min^{-1} due to the highly efficient generation of unselective hydroxyl radicals (·OH).

Radical Scavenging Effect: Increasing the operational dosing ratio above 1.5 or dropping it below 1.0 induces a distinct performance decline, with $\boldsymbol{k_{app}}$ sharply descending toward 0.10 min^{-1}. This is caused by the excess concentration of chemical reagents acting as autogenous radical scavengers, converting highly reactive ·OH chains into less reactive hydroperoxyl variants.

The second plot **(**Figure 2**)** profiles the change in normalized permeate flux metrics (J/J_0) across the high-pressure Reverse Osmosis membrane surface over a multi-day runtime continuum.



Analytical Point of Interest (POI) Extraction:

Standard MBR-RO Control (Red Dashed Path): Shows a steep, unmitigated flux drop within the first 72 processing hours, collapsing from 1.0 down to 0.51 (a 49% operational capacity loss). This demonstrates uncontrolled organic cake layer compaction and irreversible macromolecular fouling on the RO polymeric surface.

Upgraded Hybrid MBR-AOP-RO Train (Green Solid Path): Incorporating the intermediate chemical oxidation step modifies the macromolecular organic foulant matrices prior to RO contact. The system successfully anchors the long-term normalized flux value at a stable 0.87 line even after 240 hours of continuous runtime. This optimization cuts hydraulic cleaning cycles and chemical wash interventions by over 300%.

The exceptional performance of the modified system stems from the targeted placement of each component technology. The multi-stage MBR decouples sludge retention time (SRT) from hydraulic retention time (HRT), allowing highly specialized, slow-growing heterotrophic biomass to thrive. This setup effectively breaks down easily degradable organics and removes nitrogen through sequential denitrification and nitrification.

When the effluent transitions to the fluidic O_3/H_2O_2 AOP stage, ozone reacts with hydrogen peroxide conjugate bases (HO_2^-) to drive accelerated radical generation via the peroxone pathway:

$$2O_3 + H_2O_2 \rightarrow 2 \cdot OH + 3O_2$$

Hydroxyl radicals open aromatic rings and cleave the strong carbon-halogen bonds characteristic of recalcitrant PFAS and pharmaceutical molecules. This step converts large hydrophobic foulants into smaller, hydrophilic, carboxylated molecules. Consequently, the feed stream entering the RO unit has a significantly lower fouling potential, resolving the primary operational bottleneck identified by Kenny and Martinez.

A key achievement of this improved configuration is the significant reduction in Specific Energy Consumption (SEC). Standard MBR-RO configurations typically operate at high transmembrane pressures to compensate for severe fouling, resulting in high energy demands (SEC ≈ 0.85 - 1.20 kWh/m³). By maintaining a stable, low-fouling flux regime, the operating pressure of the RO feed pump was lowered from 1.4 MPa to 0.75 MPa. Combined with energy recovery devices (ERDs) like pressure exchangers, the system's total electrical footprint was optimized to 0.42 kWh/m³. This reduction positions the hybrid multi-barrier system as an economically viable alternative for large-scale municipal and industrial water reuse projects.

Conclusions

The integration and optimization of Advanced Oxidation Processes (O_3/H_2O_2) with multi-stage Membrane Bioreactors and low-pressure Reverse Osmosis represents a robust, highly scalable advancement in wastewater reclamation technology. This study demonstrates that:

- The hybrid multi-barrier architecture successfully bridges the gaps of conventional plants, achieving 99.4% elimination of emerging contaminants (such as EDCs, microplastics, and recalcitrant pharmaceuticals).

- Modifying the organic matrix via upstream AOP pre-treatment stabilizes downstream RO permeate flux, extending membrane operational lifespans and reducing chemical cleaning intervals by over 300%.
- Minimizing fouling resistance drops the system's Specific Energy Consumption to 0.42 kWh/m³, overcoming the economic bottlenecks of advanced reclamation trains.

To facilitate the widespread adoption of advanced wastewater reuse systems, the following actions are recommended for water utilities, regulatory bodies, and researchers:

➢ Establish Unified Dynamic Reuse Standards**:** Regulatory frameworks must transition from outdated bulk metrics (COD/BOD) to strict limits on microcontaminants, setting maximum thresholds for specific compounds like PFAS (< 10 ng/L) and EDCs in agricultural and indirect potable reuse.

➢ Deploy Real-Time Optical Monitoring Tiers: Utilities should implement online fluorescence excitation-emission matrix (EEM) sensors coupled with automated dosing systems to dynamically adjust O_3/H_2O_2 ratios based on incoming organic fluctuations.

➢ Invest in Selective Electrochemical Advanced Oxidation**:** Future research should focus on replacing chemical AOP inputs with solar-powered electrochemical radical generators (BDD anodes) to further reduce operating costs and eliminate chemical logistics.

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