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REVIEW PAPER

Chemical oxidation as an alternative for municipal wastewater secondary treatment: a review

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Abstract Activated sludge has been widely adopted as the cornerstone of conventional sewage treatment for over 50 years. This process can reduce biochemical oxygen demand (BOD) in wastewater and protect public health, with many systems able to remove nutrients as well. While activated sludge continues to satisfy many treatment targets, the demands on wastewater treatment are changing. There are concerns that toxic and difficult-to-degrade contaminants are contributing to environmental and human health issues. There is also increasing interest in potable reuse to strengthen water resiliency and the waste-to-resource paradigm; however, when biological secondary treatment is used, additional treatment is needed for reuse. Chemical oxidation may be an effective alternative to activated sludge to destroy difficult-to-degrade contaminants. Compared to biological systems, chemical oxidation may also be easier to operate and maintain, requiring less space for more effective treatment. This article presents a critical review of current activated sludge-based sewage treatment practices and explores the opportunity to replace biological secondary wastewater treatment with chemical oxidation. Some opportunities include the ability of chemical oxidation to degrade contaminants of emerging concern (CECs); rapid start up and shut down; and avoidance of issues associated with biological treatment such as toxic loadings, biomass washout, difficulties settling sludge, and sludge handling and disposal. This review focuses on chemical oxidation as an alternative to biological secondary treatment for municipal wastewater. Most works included in this review are referenced in Google Scholar and the Web of Science, with the majority being published between 2000 and 2023. Trends revealed include a substantial increase in investigations regarding biological treatment, but much less literature focused on chemical oxidation of municipal secondary wastewater. There were reports covering chemical oxidation for industrial wastewater and for tertiary treatment of municipal wastewater, but not for chemical oxidation as a secondary treatment method for municipal wastewater."

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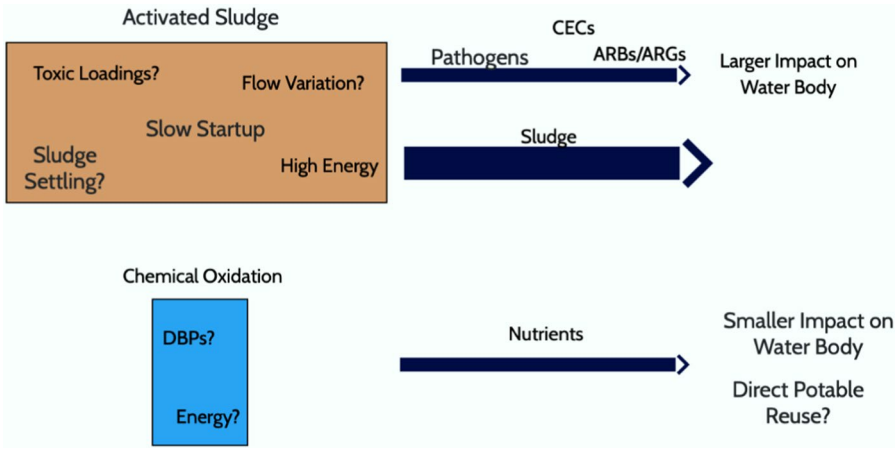
Graphical abstract



Keywords Activated sludge · Advanced oxidation · Wastewater treatment · Water reuse · Ozone

1 Current secondary treatment practices

Water resource recovery facilities (WRRFs) play a fundamental role by removing pathogens and other contaminants to protect human health and the environment. Secondary treatment is a part of many WRRFS and is defined as the removal of dissolved or suspended biodegradable organic matter from wastewater; it is assumed to involve biological treatment using microorganisms (Metcalf and Eddy 2013). Biological secondary treatment has been the cornerstone of WRRFs since the late 1800s, when biological trickling filters were introduced (Lofrano and Brown 2010). In 1913, the most widely applied secondary biological treatment process, activated sludge, was patented and began to be employed. It was widely adopted in the United States (US) after the Environmental Protection Agency's (EPA's) Clean Water Act (CWA) mandated secondary treatment in 1972 (Lofrano and Brown 2010). The adoption of activated sludge and other secondary biological treatment processes has been paramount in attaining CWA goals of fishable, boatable, and swimmable waterways in the US.

Although secondary treatment was never formally and exclusively defined as being "biological" in the CWA, secondary treatment has nevertheless become synonymous with biological treatment. Chemical oxidation processes have historically not been applied to municipal sewage for the removal of the majority of soluble and colloidal organic pollutants. It is interesting to consider why chemical secondary treatment has not been used for this application. One reason is that US secondary effluent standards established to meet the CWA were based on treatment data from publicly owned treatment works (POTWs) "practicing a combination of physical and biological treatment to remove biodegradable organics and suspended solids" (U.S. 2010). No treatment plants using chemical oxidation were included in the dataset, as they did not exist, and therefore, the option of employing chemical oxidation was not considered. In 1983, the EPA published amendments to the treatment standards that established "treatment equivalent to secondary treatment" for other biological treatment processes (e.g., trickling filters, lagoons, and oxidation ditches) that meet secondary effluent standards most of the time, avoiding massive upgrade costs to WRRFs that would otherwise need to upgrade to activated sludge (Flynn 1984). However, chemical treatment still was not considered as a potential secondary treatment equivalent.

Chemical oxidation secondary treatment may also have been essentially ignored because the United Nations defines "secondary treatment" as the "second step in most waste treatment systems during which bacteria consume the organic parts of the waste…

This is accomplished by bringing the sewage, bacteria, and oxygen together in trickling filters or within an activated sludge process" (UN. 1997). In addition, the EPA states "the *Secondary stage* of treatment removes about 85 percent of the organic matter in sewage by making use of the bacteria in it."

Although secondary biological treatment has effectively been employed to treat wastewater over the past century, it has several disadvantages. Additionally, the demands of used water recovery are changing, potentially making secondary biological treatment less appealing. For example, maintaining consistent effluent quality in activated sludge processes can be difficult. Also, transient toxic loadings to WRRFs may inhibit the microbes needed to treat water (Ren 2004). High-flow, dilute loadings from storm events in combined sewers or through inflow and infiltration can be difficult to accommodate using municipal biological treatment. These events can cause microbe washout, resulting in ineffective treatment or untreated discharges (Peters and Zitomer 2021). Additionally, compounds such as pharmaceuticals, microplastics, and other contaminants of emerging concern (CEC) may pass through activated sludge systems and be discharged into the environment via treated water or solids residual disposal (Yunlong et al. 2014). When micropollutant degradation, or potable and nonpotable water reuse are goals, tertiary treatment that employs chemical oxidation is employed to further mitigate contaminants that are not effectively removed in secondary biological treatment and to provide additional disinfection (Rizzo et al. 2019; Margot et al. 2013; Jacob et al. 2010). Multiple, previous reviews discuss the role of chemical oxidation as a pretreatment or as tertiary post treatment after biological treatment for municipal wastewater (Beltrán et al. 1997; Scott and Ollis 1995; Jeworski and Heinzle 2000; Arzate et al. 2019; Zagklis and Bampos 2022; Rout et al. 2021; Patel et al. 2021). In general, chemical oxidation has been shown to be a useful technology when paired with biological treatment for municipal wastewater. As a pretreatment method to biological treatment, chemical oxidation can partially oxidize recalcitrant compounds to form more biodegradable intermediates, thus improving overall removal. Additionally, chemical oxidation effectively removes recalcitrant compounds that remain after biological treatment of municipal wastewater. Chemical oxidation also provides disinfection beyond what conventional methods, such as chlorination, are capable of achieving, while also degrading contaminants (Galeano et al. 2019; Kokkinos et al. 2021; Yu et al. 2024).

With water reuse, it is especially interesting to consider the potential benefits of replacing the two, sequential operations of biological and chemical oxidation with a single chemical oxidation process. Previous reviews highlight the potential benefits of chemical oxidation for industrial wastewater treatment, both with and without the aid of biological treatment (Oller et al. 2011; Mantzavinos and Psillakis 2004; Amor et al. 2019; Sathasivam et al. 2019). These previous works describe chemical oxidation as a useful and effective technology to degrade recalcitrant compounds when paired with conventional biological secondary treatment of municipal wastewater (both as a pretreatment and a tertiary post treatment), and to treat recalcitrant industrial wastewater. However, no previous literature reviews describing chemical oxidation to replace biological treatment for secondary municipal wastewater were found. The information reviewed herein aims to fill the gap to describe the potential effectiveness of chemical oxidation as an alternative to conventional biological treatment for secondary municipal wastewater treatment, thus possibly elimination the need for multiple treatment steps to achieve biochemical oxygen demand (BOD) removal, recalcitrant compound degradation, and disinfection (Rizzo et al. 2019; Margot et al. 2013; Jacob et al. 2010).

This review evaluates the possibility of chemical oxidation being an alternative to conventional activated sludge and other biological secondary treatment methods. First, conventional secondary biological treatment (primarily activated sludge) advantages and challenges are reviewed. Applications for novel chemical secondary treatment processes are discussed in light of potable and nonpotable water reuse requirements.

2 Advantages and challenges of activated sludge

Activated sludge is the leading secondary treatment technology to meet CWA water quality goals. Regulatory effluent limits for total suspended solids (TSS) and biochemical oxygen demand (BOD) are established to provide sufficient water quality when paired

with disinfection (U.S. EPA 2010). Initially, activated sludge systems focused on removing BOD, followed by upgrades to remove nitrogen and phosphorus from wastewater to avoid oxygen depletion and eutrophication of receiving waters (Barnard 1975, 1974; Zitomer and Speece 1993). Activated sludge has also been shown to partially or completely degrade some CECs, while other CECs can be removed from the liquid stream by sorption and incorporation in the biosolids stream, with remaining CECs passing through untreated (Baalbaki et al. 2016).

Challenges of activated sludge operation include (1) inadequate CEC removal; (2) challenging carbon management; (3) large footprint requirement; (4) high energy demand; (5) bulking, rising, and foaming sludge issues; (6) difficulty accommodating flow variation; (7) difficulty accommodating toxic loadings; (8) slow response to changes; and (9) slow startup times. These challenges are described below.

2.1 Inadequate treatment

2.1.1 CECs

Pharmaceuticals and personal care products (PPCPs), drugs of abuse (DOA), endocrine disrupting compounds (EDCs), stimulants, and pesticides are all considered CECs that can have toxic effects in aquatic environments and are present in sewage (Blair et al. 2013; Hughes et al. 2013; Gay et al. 2016; Kidd et al. 2007; Kümmerer 2003). WRRFs have been identified as a primary source of CECs in surface waters (Yunlong et al. 2014; Purdom et al. 1994; Loos et al. 2013). Secondary biological treatment processes are only able to degrade biodegradable organic pollutants, allowing some CECs to pass through partially treated, untreated, or in the biosolids (Yunlong et al. 2014; Bolong et al. 2009; Deblonde et al. 2011).

Research surveying CECs in multiple WRRFs has shown highly variable removal, with > 80% removal of some CECs while other CECs were removed at < 25% (Yunlong et al. 2014; Verlicchi et al. 2012; Jelic et al. 2011; Tran et al. 2018). Investigation of 22 CECs at a WRRF in Canada showed that activated sludge was the main process by which CECs were removed (Baalbaki et al. 2016). Seventeen of the 22 compounds were partially degraded, six of which (ibuprofen, naproxen, amphetamine, ephedrine, dihydrocodeine, and caffeine) were > 80% removed through biodegradation (Baalbaki et al. 2016). Three of the CECs were partially removed through adsorption to biosolids (Baalbaki et al. 2016). This study's results showing partial removal of CECs by activated sludge are consistent with other research on CEC removal in WRRFs (Carballa et al. 2007; Gao et al. 2012; Joss et al. 2005). Pesticides are partially removed; for example, 60% of diuron was removed by activated sludge (Stasinakis et al. 2009). Activated sludge systems with longer solids retention times (SRTs) and nitrification were correlated with higher removal efficiencies for some CECs (e.g., antibiotics, antiphlogistics, antidepressants, and musk fragrances) possibly due to the extended treatment time (Fernandez-Fontaina et al. 2012; Suarez et al. 2010; Clara et al. 2005).

Some CECs may be removed by sorption to solids in primary and secondary treatment. Fragrance compounds and triclosan were partially removed by sorption to solids in activated sludge (< 40%), while most CECs were not significantly removed by sorption (< 5%) (Verlicchi et al. 2012; Ternes and Siegrist 2004; Yunlong et al. 2014).

Researchers also have reported biological treatment actually increases some CEC concentrations, which can be attributed to metabolites being transformed back into parent compounds (Göbel et al. 2007; Kasprzyk-Hordern et al. 2009). It is also possible that some CECs (e.g., pesticides) desorb from solid particles during biological treatment, increasing their aqueous-phase concentrations (Yunlong et al. 2014; Köck-Schulmeyer et al. 2013).

Pharmaceutical compounds have been shown to impact microbial community structure in biological secondary treatment and reduce treatment efficiency (Pires et al. 2021). Furthermore, other CECs, such as chlorinated compounds, chemical additives, and pharmaceuticals, can have a toxic effect on biological treatment processes, thus reducing efficiency (Tobajas et al. 2016; Zhao et al. 2019).

2.1.2 Antibiotic resistant bacteria (ARB) and antibiotic resistant genes (ARGs)

Overuse of antibiotics and their subsequent discharge into sewers has contributed to elevated levels of antibiotic-resistance genes and mobile genetic units in activated sludge systems (Zhang et al. 2011). Antibiotics are typically difficult to biodegrade in WRRFs,

resulting in their discharge into the environment (Verlicchi et al. 2012). The lack of CEC treatment in conventional WRRFs has also been shown to lead to antibiotic resistance in aquatic environments (Pruden et al. 2006). Additionally, ARBs and ARGs are a concern when treated wastewater and biosolids are used for agriculture and other water reuse applications (Krzeminski et al. 2019).

WRRFs can promote ARG transfer, leading to the proliferation of ARBs, especially during the activated sludge process (Zhao et al. 2019; Nguyen 2021; Yang et al. 2013). A study of 13 WRRFs that employed activated sludge with different modifications found that the percentage of ARBs in the effluent increased compared to influent, with the highest increases in facilities using anaerobic/anoxic/oxic treatment and sequencing batch reactors with long hydraulic residence times (HRTs) (Korzeniewska and Harnisz 2018).

2.1.3 PFAS

Per- and polyfluoroalkyl substances (PFAS) are another group of CECs that have gained attention due to their impacts on environmental and public health (Daly et al. 2018). PFAS have been identified in WRRF effluents, indicating that PFAS constituents are not removed (or are only partially degraded) in conventional wastewater treatment processes (Baluchová et al. 2019; Sinclair and Kannan 2006). A study of six WRRFs in China found no significant PFAS removal (yearly average), or even increased PFAS concentrations for aerobic secondary treatment systems (Chen et al. 2018). Negative removal in aerobic biological systems can be attributed to the oxidation of precursors to perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), thereby increasing their concentrations in the effluent (Chen et al. 2018; Guerra et al. 2014; Zhang et al. 2013).

2.2 Carbon management

Generation of waste solids (i.e., waste activated sludge) from biological treatment processes poses challenges with stabilization and disposal/reuse. Sewage sludge solids stabilization and disposal can make up 50–60% of WRRF operating costs (Coma et al. 2013; Pilli et al. 2015). Typically, aerobic or anaerobic digestion is employed to stabilize and reduce the volume of sewage sludge; anaerobic digestion also generates biomethane for heat and electrical energy generation (Eddy 2013). Secondary biological treatment not only results in the generation of a significant mass of sludge that must be stabilized and disposed of but also results in sludge that is more difficult to digest than primary sludge (Borzooei et al. 2019). Waste activated sludge (WAS) has limited methane potential compared to primary sludge (Grübel and Suschka 2015). Minimizing or eliminating WAS production and subsequent sludge handling can greatly reduce WRRF operating costs and environmental impacts (Coma et al. 2013; Banti et al. 2020).

Stabilized sludge must be disposed of or reused. This can be done through appropriate landfilling, land application, composting, incineration, or other processes. Sludge handling and disposal is an expensive endeavor, with an average cost of $65/wet ton (Smith 2020). Disposal fees for landfilling biosolids add to WRRF operational expense. Land application is often considered to be more favorable, as nutrients in the biosolids can be beneficial to agriculture and may lead to reduced disposal costs compared to landfilling (Lu et al. 2012).

However, land application of biosolids can introduce several challenges. Heavy metals in biosolids can accumulate in soils, damaging crops, leaching into groundwater, and bioaccumulating in animals and humans (McGrath et al. 1994). Biosolids can also be a source of pathogenic contamination and antibiotic resistance (Krzeminski et al. 2019; Pritchard et al. 2010; Viau et al. 2011; Brooks et al. 2007). PFAS have been shown to sorb to sewage sludge, exiting WRRFs through a solids pathway in addition to the liquid effluent (Sinclair and Kannan 2006; Arvaniti et al. 2012, 2014; Higgins et al. 2005; Ochoa-Herrera and Sierra-Alvarez 2008). PFAS homologs are only partially degraded in sludge handling processes such as anaerobic digestion, leading to the spread of PFAS in the environment and possible uptake into crops if biosolids are land applied (Lakshminarasimman et al. 2021; Sepulvado et al. 2011). Eutrophication is also a concern, leading to the adoption of nutrient management plans and possible recommendations for reduced biosolids nutrient application to farmland (Cherry et al. 2008). Land application, especially near densely populated urban areas, has steadily declined due to increasing regulations, competitive landfilling

rates, contamination liability concerns, and public scrutiny (Smith 2020).

2.3 Large footprint requirement

Secondary biological treatment systems, especially activated sludge, take up a large amount of space. Activated sludge systems have long HRTs, increasing with the level of treatment (e.g., nutrient removal), ranging from 4 to 8 h or more for conventional treatment to 20–40 h for extended aeration treatment (Eddy 2013). These long HRTs lead to large aeration tanks and secondary clarifiers, with large footprints and increased capital costs. Beyond the liquid stream treatment, large volumetric flows of WAS are produced with activated sludge, requiring equipment and space to handle WAS before disposal.

2.4 Energy demand

Reducing energy demand for wastewater treatment is a high priority to help reduce costs and greenhouse gas emissions. WRRFs are estimated to consume 0.8% of US energy, costing about $2 billion per year (Lemar and Fontaine 2017; Electric Power Research Institute 2013). It is estimated that WRRFs constitute over 20% of the energy demand for municipal public utilities (Means 2004). Of this, approximately 60% of conventional WRRF energy demand is for the aeration of activated sludge (Gikas 2017; Shi 2011; Svardal and Kroiss 2011). This high energy demand can be a problem for small communities that do not have the financial base to operate an activated sludge process, especially one that can remove nutrients. The high energy demand also contributes to greenhouse gas emissions and climate change if fossil fuels are used for energy production (Banti et al. 2020).

2.5 Bulking, rising, and foaming sludge issues

Issues with solids separation in activated sludge systems can reduce treatment efficiency and capacity. Bulking sludge, rising sludge, and foaming are the most common operational issues in an activated sludge plant that may interfere with, or even halt, BOD and nutrient removal (Eddy 2013). Bulking sludge has poor settling characteristics and can result in high effluent suspended solids and reduced treatment performance since it can reduce the amount of active biomass in the system (Nittami and Batinovic 2021). Rising sludge, although rare, can occur when denitrification takes place in secondary clarifiers.

Foaming problems are typically associated with diffused aeration systems and can also lead to foaming issues in subsequent digestion processes (Westlund et al. 1998). Operational changes (e.g., reduced sludge age and use of selectors), chemical oxidizing agents (e.g., chlorine), and coagulants/polymers (e.g., cationic polymer) may effectively suppress foaming (Hwang and Tanaka 1998; Jenkins et al. 2003; Tsang et al. 2008; Pal et al. 2014).

2.6 Difficulty accommodating flow variation and toxic loadings

Changing flows and the associated changes in loadings can greatly impact activated sludge performance. Reduced flows due to droughts can result in higher pollutant concentrations, while increased flows due to storm events and inflow and infiltration can hydraulically overload activated sludge facilities, resulting in biomass washout and poor treatment.

As droughts increase in frequency and duration, there is a push toward water conservation to reduce demand on water supply systems (Dinar and Schwabe 2015; Baerenklau et al. 2014). Without reducing the mass of pollutant discharge, reducing water flows through methods such as low-flow fixtures and reducing the duration of water use (e.g., shorter showers, turning off the faucet when not using) increases the concentration of pollutants reaching WRRFs. Increased loadings of inorganic salts (e.g., NaCl) can reduce oxygen transfer and uptake rate as well as total organic carbon (TOC) removal in activated sludge systems (Wang et al. 2005).

Increased dilute flows from storm events and seasonal variations also influence activated sludge process treatment efficiency for combined sewer systems and sanitary sewers with infiltration and inflow. One common practice in the US is to store excess flows during storm events and subsequently treat the stored water when flows are lower (Peters and Zitomer 2021). High flow variation is unfavorable since increased hydraulic loading can dilute biomass in the activated sludge process, and can result in microbial community washout, thereby decreasing treatment efficiency (Gaudy and Engelbrecht 1961). Storm events can decrease nitrification, nitrogen removal,

and particle separation efficiency in secondary treatment (Wilén et al. 2006). Langeveld et al. (2013) studied rainfall impacts on a WRRF in Eindhoven, The Netherlands. After a dry period of 38 days, a relatively small event occurred that led to multiple combined sewer overflows and a 5-week loss of activated sludge hydraulic capacity. The effluent quality never exceeded permit limits, but influent pumping had to be reduced, resulting in more CSO water being released untreated to receiving streams. In Oslo, Norway, temporary snow melting periods created high flow conditions of cold influent at the WRRF, resulting in reduced biological nitrogen removal and a decrease in secondary clarifier solids removal efficiency (Plósz et al. 2009). Biological nitrogen removal is of particular concern in areas affected by cold temperatures, as nitrifying organisms are slow-growing and highly sensitive to low temperature (Eddy 2013).

Toxic loadings can be introduced to wastewater streams as "first flushes" from wet weather events and other discrete industrial discharges. These shock loads can negatively impact the metabolic processes and microbial communities in biological treatment systems (Gaudy and Engelbrecht 1961; Krishnan and Gaudy 1976).

2.7 Slow response to changes

Secondary biological treatment has relatively slow response times to changing loadings and changing aeration inputs due to the time required for microbial growth. In particular, activated sludge processes are difficult to control as they are complex systems exhibiting nonlinear behavior (Holenda et al. 2008). Additionally, activated sludge systems are difficult to monitor with sensors due to sensor fouling and calibration issues; therefore, lab testing is often required, further increasing response time. On-line monitoring of activated sludge processes has been studied extensively, with sensors designed to reduce response time through consistent measurements of parameters such as dissolved oxygen, ammonia, and nitrate concentrations (Gernaey et al. 2001; Liu et al. 2000). Activated sludge modeling is also used to better predict and manage these systems to increase the consistency of effluent quality and improve energy efficiency (Newhart et al. 2019; O'Brien et al. 2011; Stare et al. 2007). Although sensors and modeling strategies can help, they are not always reliable and can be difficult to manage with the activated sludge slow response time to variable wastewater influent quantity and quality.

2.8 Slow startup times

Biological secondary treatment systems cannot be started immediately, as time is needed for biomass to establish and acclimate to a specific wastewater. Seeding from other basins can reduce startup times, provided biomass is readily available. Typical startup times for conventional activated sludge range from 20 to 60 days if the inoculum biomass is adapted to the wastewater and sufficient biomass is available for inoculation (Pronk et al. 2015). Startup times can be longer if biomass inoculation is not used. Although long startup times are not an issue for continuous operation of biological treatment, they do present a challenge when a system must be taken offline for maintenance and then restarted.

3 Chemical oxidation

Activated sludge has been the subject of continuous improvements over more than 100 years. However, there is less work developing potential, alternative secondary treatment technologies, such as chemical oxidation. There have been a limited number of publications discussing chemical treatment of sewage since 1990, although there is an increasing trend (Fig. 1). In comparison, a Web of Science search of "activated sludge" returned over 2,600 results between January and October 2022. While chemical oxidation is not typically used for secondary treatment of municipal sewage, it may offer multiple advantages that substantiate its consideration as an activated sludge alternative. A Venn diagram is included in (Fig. 2) to highlight abilities for both secondary biological and chemical treatment.

Because of its ability to degrade CECs, chemical oxidation has been used to treat water with low concentrations of organic constituents, including in-situ groundwater, drinking water, tertiary wastewater, and high-strength wastes that are difficult to treat using biological systems, such as landfill leachate and industrial wastewater (Seol et al. 2003; Hodaifa et al. 2019; Wiszniowski et al. 2006; Gomes et al. 2017;

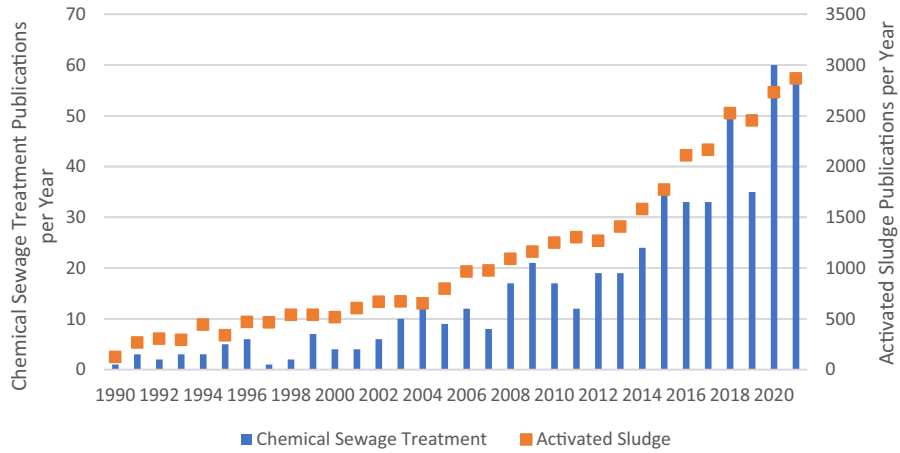


Fig. 1 Annual number of publications regarding chemical treatment of sewage and activated sludge. The Web of Science search was performed on October 20, 2022, using keywords "Chemical Sewage Treatment" or "Activated Sludge" for the period 1990–2021

Bhatti et al. 2011). Chemical oxidation systems can have faster reaction rates than biological treatment, faster response time to changes in influent characteristics, do not need time to develop biomass, and may accommodate fluctuating flows and loadings more easily than biological systems (Ikehata et al. 2006). Unlike secondary biological treatment, chemical oxidation systems do not generate WAS, thus eliminating the need for sludge handling from the secondary treatment step. Common chemical oxidants used in water and wastewater treatment for the removal of contaminants include ozone and advanced oxidation processes (AOPs). Table 1 shows a summary of the advantages and disadvantages of activated sludge and chemical oxidation treatment methods.

3.1 Ozone

Ozone is a strong oxidant (redox potential of 2.07 V), making it desirable for trace organic removal and disinfection of water in one treatment step (Wang and Chen 2020). Ozone may react with constituents in water through both direct (ozone-based) and indirect (hydroxyl radical [HO·]-based) pathways (Wang and Chen 2020).

Ozone's ability to oxidize trace organics such as CECs has led to its adoption in drinking water and tertiary wastewater treatment. Ozone is effective at degrading hydrophobic CECs that would otherwise not be removed by conventional treatment. For example, Rosal et al. (2010) showed that fifteen minutes

Table 1 Advantages and disadvantages of activated sludge and common advantages and disadvantages of chemical oxidation treatment

	Activated sludge	Chemical oxidation
Advantages	• Established secondary treatment technology • Effective removal of BOD • Effective removal of nutrients	• Can degrade CECs • Provides disinfection • Fast startup and response times • Relatively small footprint • No sludge handling and disposal • Can integrate well into potable reuse facilities
Disadvantages	• Ineffective CEC removal • Treatment upsets by toxic loadings, flow variations • Large space and energy requirements • Need for sludge handling and disposal • Sludge bulking and settling issues	• Not able to remove nutrients on its own • No established kinetic values for secondary municipal wastewater • Need for post treatment to handle toxic byproducts and disinfection byproducts (DBPs) • May be energy intensive

of ozonation at a dose of 50—220 mM after secondary biological treatment degraded 37 of the 54 CECs studied to below the level of quantification (Rosal et al. 2010). Ozone is effective at removing most antibiotics, beta-blockers, hormones, and contraceptives, with mixed results for non-steroidal anti-inflammatory drugs (NSAIDs) and anti-anxiety medications. However, ozone is not very effective for oxidizing lipid regulators and X-ray contrast media (Ikehata et al. 2006).

Ozone can inactivate pathogens including viruses, bacteria, fungi, spores, protozoa, nematodes, and algae (Rojas-Valencia 2011). In particular, ozone can inactivate microbes that may be chlorine- and/or UV-resistant. For example, ozone is more effective at completely inactivating *Cryptosporidium* oocysts compared to chlorine dioxide, chlorine, and monochloramine (Peeters et al. 1989; Korich et al. 1990). Chlorine-resistant bacteria are also a source of concern due to the widespread use of chlorine disinfection. Ozone has been shown to inactivate these organisms and is capable of destroying chlorine-resistant genes (Ding et al. 2019). Ozone was effective for the destruction of ARBs in a full-scale WRRF, but only at doses higher than required for chemical CEC removal (Czekalski et al. 2016).

Challenges associated with ozonation include the fact that partial oxidation of pollutants by ozone may increase toxicity and could produce more difficult to degrade intermediates (Wang and Chen 2020). Ozone also has low solubility in water and is energy-intensive to generate (Wang and Chen 2020). Finally, ozonation of waters containing bromate could lead to the formation of brominated disinfection byproducts (DBPs) (Wang and Chen 2020; Beltrán et al. 2021).

3.2 Advanced oxidation processes (AOPs)

AOPs come in many different configurations and can overcome challenges with other chemical and biological oxidation processes. AOPs are chemical oxidation methods that typically rely on HO·, which are highly reactive (redox potential of 2.80 V), non-selective oxidants (Wang and Chen 2020). AOPs may also feature other radicals, such as sulfate radicals or superoxide radicals. AOPs can be categorized as ozone-based, UV-based, catalytic, physical, electrochemical, and non-HO· forming AOPs, or combinations thereof (Miklos et al. 2018). A summary of different types of AOPs is shown in Table 2.

When comparing AOPs, it is important to account for the energy efficiency, commonly assessed as the electrical energy needed to reduce the concentration of a contaminant by an order of magnitude, or the electrical energy per order (EEO). AOPs have been grouped into those with EEOs < 1 kWh m^{-3} (e.g., O_3-based AOPs, UV/H_2O_2), 1–100 kWh m^{-3} (e.g., photo-Fenton), and > 100 kWh m^{-3} (e.g., UV photocatalysts) (Miklos et al. 2018). It should be noted that these broad classifications include data for AOP treatment of a wide range of different target contaminants (Miklos et al. 2018).

Ozone alone is considered an AOP or AOP-like process as ozone can break down into HO· when reacting with hydroxide ions or organic matter (Miklos et al. 2018; Merényi et al. 2010a, 2010b; Buffle and Gunten 2006). Ozone-based AOPs enhance the breakdown of ozone to HO·, which can be accomplished homogenously (elevated pH, O_3/H_2O_2); heterogeneously with metal oxides, activated carbon, or other catalysts; or photocatalytically with UV (Miklos et al. 2018). The non-selective nature of HO· allows AOPs to readily degrade organics, with much higher kinetic constants than ozone alone ($k_{HO·}$ of 10 (Peters and Zitomer 2021)–10 (Rizzo et al. 2019) M^{-1} S^{-1}, k_{O3} of 10^{-3}–10 (Eddy 2013) M^{-1} S^{-1}) (Legube and Karpel Vel Leitner 1999). Ozone-based AOPs overcome the selective nature of ozone, increasing the range of contaminants degraded and the kinetics of degradation by leveraging both direct ozonation and HO· (Miklos et al. 2018). Degradation of natural organic matter (NOM) and effluent organic matter (EfOM) by AOPs has been shown to reduce DBP formation compared to ozonation alone (Beltrán et al. 2021; Lamsal et al. 2011).

Although ozone-based AOPs alleviate some challenges of conventional ozonation, other challenges still exist. Use of ozone-based AOPs to treat waters with > 100 µg/L Br^- can result in 5–50% conversion of Br^- to the undesirable DBP BrO_3^- from direct ozonation (Hübner et al. 2015; Gunten 2003). The O_3/H_2O_2 process may provide a limited benefit through the efficient breakdown of ozone in wastewaters and the avoidance of scavenging reactions, but it also may require subsequent removal of excess H_2O_2 (Miklos et al. 2018; Hübner et al. 2015). Using catalytic ozonation can have issues

Table 2 Advantages and disadvantages of different types of AOPs

Type of AOP	Advantages	Disadvantages	References
Ozone-based	• Reduced energy for O_3 production compared to ozonation alone • Reduced bromate and other DBP formation compared to ozonation alone • No waste disposal • Use both ozone and HO· for contaminant destruction and disinfection	• Can produce bromate • Catalytic ozonation can have water/catalyst contact issues	Beltrán et al. (2021), Miklos et al. (2018), Lamsal et al. (2011), Hübner et al. (2015), Huber et al. (2003)
UV-based (ie, photocatalytic, UV-H_2O_2	• No waste disposal • No production of halogenated DBPs • Both UV and HO· provide disinfection • No residual catalyst separation and handling	• High dissolved organic matter (DOM) concentrations can interfere with UV • Catalyst separation and recovery needed for photocatalytic systems • DBP formation concerns for high natural organic matter (NOM) concentrations	Miklos et al. (2018), Buchanan et al. (2006)
Fenton and Fenton-like	• Catalysts can be separated by magnets • Considered low cost	• Require low pH (pH = 3) • Must dispose of metal sludge	Sánchez Pérez, et al. (2013), Brienza and Katsoyiannis (2017)
Physical (ie, microwave, cavitation, etc.)	• No DBP formation concerns • No waste disposal • No additional chemicals needed	• High energy costs	Miklos et al. (2018), Goel et al. (2004), Mahamuni and Adewuyi (2010)
Electrochemical	• No waste disposal • No additional chemicals needed	• Limited by diffusive transport of radicals • Possibility to form DBPs at electrode surface	Miklos et al. (2018), Kapałka et al. (2009)
Sulfate radicals	• Impacted by scavenging less than HO·	• Lower redox potential than HO·	Brienza and Katsoyiannis (2017), Mahdi Ahmed et al. (2012)

with mixing, as HO· are generated at the catalyst surface and do not diffuse far into bulk solution before degrading.

UV-based AOPs rely on UV irradiation combined with radical-promoting mechanisms. UV-based AOPs include UV/H_2O_2, UV/TiO_2, and the UV/Fenton process (Miklos et al. 2018). Full-scale UV/H_2O_2 operations have been implemented for potable reuse and surface water treatment (Audenaert et al. 2011; Kruithof et al. 2007). The formation of chlorate, perchlorate, and bromate is not a concern in UV-based AOPs (Miklos et al. 2018). Additionally, the UV fluences for AOPs typically exceed those needed for 4-log inactivation of most pathogens, thus providing UV disinfection with AOP treatment (National Primary Drinking Water Regulations: Long Term 1 Enhanced Surface Water Treatment Rule. Final rule 2006; Miklos et al. 2018).

Challenges associated with UV-based AOPs include difficulty when waters contain high DOM concentrations (> 10 mg-C/L); additionally, inorganic byproducts can form when irradiated with vacuum or low-pressure UV (Buchanan et al. 2006). UV irradiation of nitrate can also form nitrite at wavelengths below 240 nm, which may be an issue when using vacuum or medium-pressure UV, although most UV systems are currently low-pressure UV (Sharpless and Linden 2001). Nitrate oxidation by medium-pressure UV can result in the formation of potentially mutagenic organic DBPs in the presence of DOM (Hofman-Caris et al. 2015; Kolkman et al. 2015; Martijn et al. 2014). For photocatalytic AOPs (such as UV/

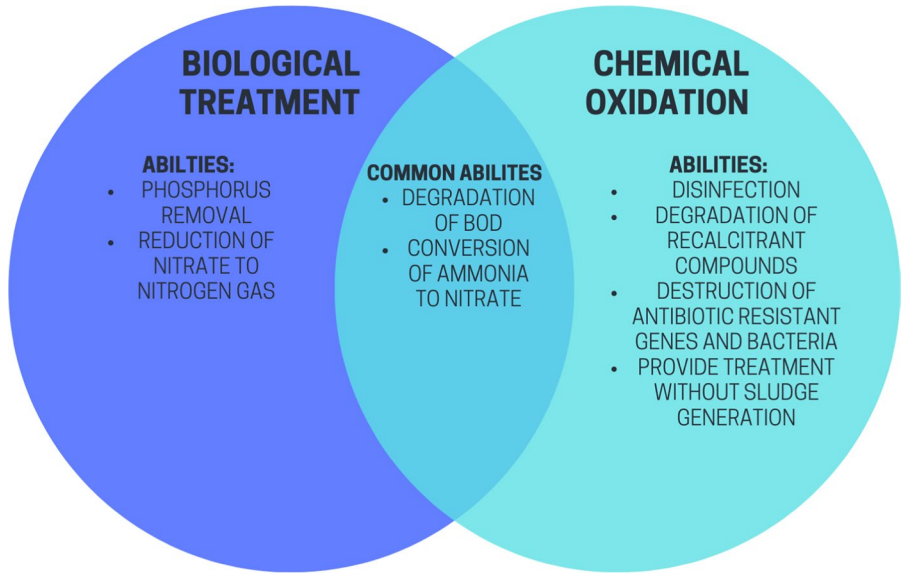


Fig. 2 Advantages of biological treatment and chemical oxidation for secondary treatment of municipal wastewater

TiO_2), separation of the catalyst from the bulk solution is an important design consideration. In instances where catalysts are immobilized, diffusion limitations can impede process efficacy, similar to ozone-based AOPs.

The Fenton process generates HO· with ferrous iron and H_2O_2 in acidic conditions (pH = 3) (Wadley and Waite 2004). Fenton-like processes substitute ferrous iron for other materials to activate H_2O_2 (Bokare and Choi 2014). Fenton and Fenton-like processes are considered low-cost AOPs, and the use of iron and other magnetic metals facilitates separation with magnets (Sánchez Pérez, et al. 2013). However, operation at low pH to prevent metal precipitation requires subsequent pH adjustment prior to discharge (Wadley and Waite 2004). Additionally, iron/metal sludge is generated and must be disposed of in these processes (Brienza and Katsoyiannis 2017).

Physical AOPs involve the manipulation of the water matrix to produce HO· instead of relying on chemical addition. These processes include plasma, ultrasound, and microwave (Miklos et al. 2018). Plasma AOPs use strong electric fields to initiate physical and chemical reactions (e.g., direct oxidation, radical generation, and shock waves) to degrade contaminants (Bo et al. 2014; Locke et al. 2006; Hijosa-Valsero et al. 2014). Ultrasound AOPs use ultrasonic waves (20–500 kHz) to form microbubbles that then collapse, generating high temperature (> 5000 K), high pressure (> 1000 bar), and highly reactive radicals that degrade contaminants through thermal decomposition and radical reactions (Miklos et al. 2018; Mason and Pétrier 2004). Microwave AOPs use high-energy radiation (300 MHz–300 GHz) to oxidize contaminants, often paired with UV, oxidants, or catalysts (Han 2004; Zhihui et al. 2005; Bo et al. 2006). Physical AOPs suffer from high energy costs, leading to the investigation of hybrid systems (e.g., combined with UV, oxidants) (Goel et al. 2004; Mahamuni and Adewuyi 2010).

Electrochemical AOPs use an electrode (often boron-doped diamond for electrooxidation processes) to generate HO· directly from water oxidation (Chaplin et al. 2013; Chaplin 2014). Electrochemical AOPs can generate radicals without chemical additives and are viewed as eco-friendly compared to other AOPs (Miklos et al. 2018). However, HO· generation happens at the surface of the electrode and diffusion is limited to about 1 µm, making diffusive transport the limiting mechanism for oxidation efficiency (Miklos et al. 2018; Kapałka et al. 2009). High radical densities at the electrode surface can also form chlorate, perchlorate, bromate, and other oxidation byproducts (Bergmann and Rollin 2007).

Beyond HO·, sulfate radicals can also be generated in AOPs. Sulfate radicals are strong oxidizers (redox potential of 2.60 V) and react through the one-electron oxidation mechanism, which reduces the impact of organic and inorganic scavenging (Brienza and Katsoyiannis 2017; Mahdi Ahmed et al. 2012).

Sulfate radicals are generated by activating precursors such as potassium persulfate or peroxymonosulfate salts through similar methods as other AOPs (Fenton/Fenton-like process, photocatalytic and mechanical activation) (Brienza and Katsoyiannis 2017). Sulfate radical AOPs can effectively degrade recalcitrant contaminants such as hormones, pharmaceuticals, and pesticides (Brienza et al. 2014; Zhang et al. 2015; Ahmed et al. 2014).

Compared to biological and other chemical oxidation methods, AOPs have been shown to degrade many more recalcitrant organic compounds and DBP precursors. Reviews of AOPs have reported effective degradation of phenols, pesticides, dyes, pharmaceuticals, EfOM, NOM, and many CECs (Wang and Chen 2020; Deng and Zhao 2015; Babu et al. 2019; Salimi 2017).

AOPs also provide disinfection and can destroy ARBs and ARGs. Radicals such as HO· have been shown to destroy cell envelopes, enzymes, proteins, lipids, nucleic acids, and intracellular substances, resulting in inactivation (Duan et al. 2021; Kokkinos et al. 2021). AOPs with UV irradiation have also been shown to have a synergistic effect, providing greater pathogen inactivation (Sgroi et al. 2021). Zhang et al. (2016) compared Fenton and UV/H_2O_2 and found 1.55–3.78 log removal of four target ARGs (*sul1, tetX, tetG,* and *intI1*) using both systems.

Challenges with AOPs involve the need to overcome scavenging, possible formation of toxic intermediate compounds and DBPs, and high cost of treatment. Reactions with non-target constituents, or HO· scavengers, can prevent radicals from breaking down the target contaminants. Scavengers include carbonate and bicarbonate, chloride, and NOM/EfoM (Nakatani et al. 2007; Grant and Hofmann 2016). The incomplete oxidation of organics can also lead to increased toxicity from AOPs (Babu et al. 2019). Finally, although less prevalent than chlorine or ozone DBP formation, AOPs may still form DBPs depending on the type of AOP, reaction time, and water matrix composition (Lamsal et al. 2011). Pure HO· has been shown to react with bromide (Br^-), forming bromate (BrO_3^-)$^-$, although bromate formation is hindered by dissolved organic matter (DOM) and in processes with excess H_2O_2 (Gunten and Oliveras 1998; Lutze et al. 2014; Von 2003). Additionally, if oxidative chlorine species are abundant (e.g., ClO·, OCl^-), oxidation by HO· to chlorate and perchlorate is possible (Miklos et al. 2018). AOPs are also typically energy-intensive, leading researchers to compare different AOPs and EEO values to determine economic feasibility in relation to treatment performance (Sgroi et al. 2021).

3.3 Challenges with chemical oxidation

Chemical oxidation as a secondary treatment method is not without its challenges. In addition to the individual challenges with different types of chemical oxidation, there are also challenges inherent to chemical oxidation as a whole. These challenges include energy usage and the need for post treatment.

The perceived high energy demand for chemical oxidation is one of the major hurdles to its adoption as a secondary treatment method (Miklos et al. 2018). Ozone generation requires energy in addition to the energy needed to transfer ozone into water, similar to the energy required to transfer oxygen to water for activated sludge treatment. Many AOPs require energy for UV light, ozone, H_2O_2 addition, or combinations thereof. Without establishing the kinetics of chemical oxidation secondary treatment, it is unknown if the high energy demand can be offset by savings compared to activated sludge, such as decreased solids processing and detention times.

Another concern with chemical oxidation is the potential need for post treatment. Chemical oxidation methods may generate toxic byproducts and DBPs that need to be removed (Lamsal et al. 2011; Babu et al. 2019). Tertiary treatment processes (e.g., activated carbon, membrane filtration) may be required to mitigate DBPs and reduce toxicity (Toor and Mohseni 2007; Listiarini et al. 2010). Selection of the appropriate chemical oxidation method will depend on influent characteristics, requiring pilot studies to evaluate which method is best, what HRT is required, and what post treatment processes are needed to ensure effluent water quality.

Additionally, chemical oxidation typically cannot remove nutrients, so another process will be needed. Ozone can directly oxidize ammonia to nitrate, which can be removed by reverse osmosis, whereas ammonia is not appreciably removed by reverse osmosis (Schoeman and Steyn 2003; Krisbiantoro et al. 2020). A UV/H_2O_2 AOP was also shown to convert up to 38% of soluble non-reactive phosphorus to soluble reactive phosphorus, which could be removed

by subsequent treatment processes (Venkiteshwaran 2021). This is beneficial as effluent phosphorus limits are becoming more stringent, with some limits below 0.10 mg/L total phosphorus so that even low concentrations of nonreactive phosphorus in sewage can impact effluent permits. Additionally, a photocatalytic AOP (UV/TiO_2) coupled with ultrafiltration removed 90–97% of total phosphorus from municipal wastewater effluent (Gray et al. 2020).

4 Potential additional applications for chemical oxidation secondary treatment

Advantages of chemical oxidation include the ability to reach advanced treatment goals (CEC destruction and pathogen inactivation), relevance for decentralized treatment (smaller footprint, possible lower energy demand, no secondary sludge, and fast response and startup times), its ability to handle flow variations, and its ability to be used for water reuse applications. These aspects are considered in the following sections.

4.1 Advanced treatment goals

As treatment of CECs and ARB/ARGs is increasingly considered, alternatives to secondary biological treatment will be needed. Chemical oxidation can degrade CECs and ARB/ARGs (Wang and Chen 2020; Deng and Zhao 2015; Babu et al. 2019; Salimi 2017; Duan et al. 2021; Kokkinos et al. 2021), whereas many CECs are not removed in biological treatment and ARB/ARGs can increase in biological treatment systems (Yunlong et al. 2014; Bolong et al. 2009; Deblonde et al. 2011; Nguyen 2021; Yang et al. 2013). Tertiary chemical oxidation systems are already in use to degrade contaminants that are not removed during biological treatment (Kharel 2020; Piras 2020).

Although tertiary chemical treatment can improve effluent water quality, it cannot remove CECs and ARB/ARGs in biosolids. The spread of CECs such as PFAS and ARB/ARGs through biosolids is of increasing concern for agriculture (Krzeminski et al. 2019; Lakshminarasimman et al. 2021; Sepulvado et al. 2011). Replacing biological secondary treatment with chemical treatment would allow for the degradation of CECs and ARB/ARGs in the liquid stream while avoiding the production of biosolids and thus the spread of CECs and ARB/ARGs through land application.

4.2 Decentralized treatment

Because of short detention times, lack of biosolids production, and integration of contaminant destruction and disinfection, chemical secondary treatment systems are candidates for decentralized treatment systems. Decentralized systems typically must be small due to their application (e.g., within buildings, in remote communities, rest stops and parks, mobile/disaster relief, expeditionary military applications, etc.) and often do not have full-time trained operators nor conveyance networks to bring water to them (Hur et al. 2023). Chemical systems require a smaller footprint as they may operate at lower HRTs than biological systems, and do not need a separate treatment process for disinfection. They also avoid the infrastructure needed to stabilize, store, and dispose of WAS. Additionally, there may be less operator attention required for chemical oxidation systems since there is no need to manage biological growth.

Chemical systems can also be designed to minimize onsite chemical storage. Ozone-based systems can be operated with little to no chemical storage, either using liquid oxygen or ambient air with an onsite oxygen concentrator. Some AOPs can operate with ozone and/or UV bulbs, with no additional chemical storage.

Another benefit of chemical treatment systems is the ability to allow the system to remain idle, only operating when needed (Peters and Zitomer 2021). Because chemical treatment systems do not need time for bacterial growth and acclimation, they can be started and shut down as needed. This makes chemical secondary treatment suitable for areas with seasonal tourism or for disaster relief, which only require extra treatment capacity for short periods.

4.3 Accommodation of wet weather/dilute high flows

High flow events from wet weather, especially in combined sewer systems, are challenging for biological treatment systems, but are candidates for chemical oxidation. Chemical treatment systems do not have issues with biomass washout, allowing them to maintain treatment efficiency during high-flow

events. They also can start operation rapidly, allowing for a quick response time if a WRRF is experiencing transient, high flows. Chemical oxidation systems have also been proposed for the "peak plant" concept, which is a facility that remains idle until it is needed to treat excess flow (Peters and Zitomer 2021).

5 Highlighted benefits of chemical oxidation for integrated water reuse

Increasing water demands from population growth and economic development paired with increasing occurrences and intensities of droughts from climate change make water scarcity a high-impact risk (Liu et al. 2017; WEF 2015). An estimated four billion people face water scarcity at least one month each year (Mekonnen and Hoekstra 2016). Water scarcity's effects spread outside of areas that are directly impacted by the lack of water. Agriculture is one of the largest water-consuming sectors, and water scarcity leads to global challenges in food production (Mancosu et al. 2015).

Potable water reuse is a technological solution that is receiving increased attention to combat challenges related to water scarcity. Potable water reuse falls into two categories, indirect potable reuse (IPR) and direct potable reuse (DPR). IPR systems have an environmental buffer (e.g., lake, groundwater aquifer) between wastewater and drinking water treatment, while DPR systems do not (Jeffrey et al. 2022). The pros and cons of IPR and DPR are discussed elsewhere (Dow et al. 2019; Herman et al. 2017).

The two main technological challenges with potable water reuse involve pathogen inactivation and the removal of CECs (Jeffrey et al. 2022). Although IPR and DPR systems are effectively able to eliminate these concerns, issues arise with process upsets and failures, which could result in the discharge of pathogens, such as viruses and *Cryptosporidium parvum,* and CECs into the environmental buffer or distribution network (Jeffrey et al. 2022). As such, a multi-barrier approach is taken, which provides treatment redundancy in case of upsets or failure of individual unit operations (e.g., UV-AOP following reverse osmosis) (Lahnsteiner et al. 2018). Chemical oxidation (e.g., AOPs, O_3-biological activated carbon, ozone for membrane biofouling control) is frequently applied to potable reuse applications for its ability to degrade CECs and inactivate pathogens (Jeffrey et al. 2022; Gerrity et al. 2014; Stanford et al. 2011). Potable reuse treatment systems typically involve WRRF effluent going to an advanced water treatment plant (which houses additional unit operations to create potable water) before distribution or storage in an environmental buffer (Gerrity et al. 2013). By replacing secondary biological treatment at the WRRF with chemical treatment, the WRRF and advanced water treatment plant could be integrated to reduce treatment time, complexity, and cost. Figure 3 shows a typical IPR treatment system, while Fig. 4 shows a proposed integrated DPR treatment system.

The paradigm shift from "wastewater" and "drinking water" to "water" is beneficial in many instances when considering water scarcity issues. Integrating WRRFs and drinking water plants would eliminate pollution from wastewater discharges, reduce groundwater and surface water demands, and provide another reliable supply of clean water (the only supply increasing in quantity). This would also allow a single managing entity to control the water treatment system, allowing for increased efficiency.

Integrated water recovery facilities will require advanced unit operations compared to conventional wastewater and drinking water treatment. Advanced primary treatment (e.g., chemically enhanced primary treatment (CEPT) or cloth media filtration (CMF)) could be implemented to reduce the loading on secondary treatment systems, thus improving their efficiency. Chemical treatment instead of biological treatment with disinfection could provide the necessary CEC and pathogen mitigation while also avoiding issues associated with sludge handling. Additionally, chemical secondary treatment pairs well with membrane filtration (Fig. 4), which is often used in potable reuse applications, as it can prevent biofouling, thereby reducing the energy and cleaning needed for membrane filtration (Stanford et al. 2011).

6 Future research needs

Before chemical oxidation is applied for secondary treatment of sewage, some hurdles must be overcome. These include evaluation of energy requirements, kinetic studies of secondary chemical oxidation of sewage, and nutrient conversion and removal.

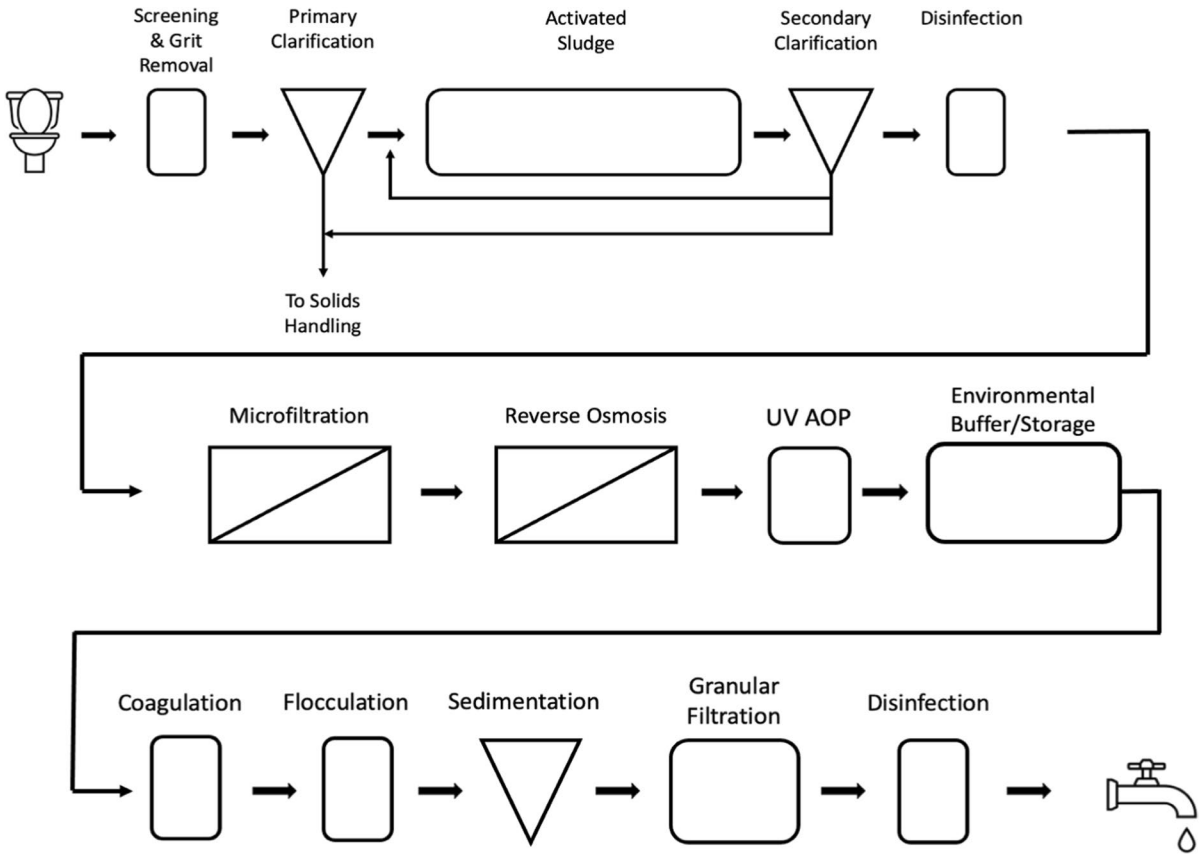


Fig. 3 Typical IPR treatment system

Additionally, there are improvements to be made for biological secondary treatment methods as well. Some of these include improved understanding of microbial communities, improving modeling for biological reactors, advanced reactor technologies (sequencing batch reactors, UNITANK, aerobic and anerobic membrane bioreactors, etc.), and biological methods for CEC removal, which are discussed in other literature (Loosdrecht et al. 2015; Cecconet et al. 2017; Abdelrahman et al. 2021; Tran 2022; Liu et al. 2022).

6.1 Energy requirements

Energy requirements for chemical oxidation systems are one of the largest hurdles to their adoption. Optimal process selection and methods to increase energy efficiency are imperative for chemical oxidation systems to be cost-competitive. Current research is focused on reducing energy costs or impacts, including through the use of sustainable energy sources, catalysts, membrane-based AOPs, sulfate-based AOPs and artificial neural network (ANN) process optimization (Giwa et al. 2021). Using ANN to model the kinetics of contaminant removal with a photochemical AOP reduced energy requirements and treatment cost (Göb et al. 1999). ANN may improve understanding of degradation kinetics when applying AOPs to a complex matrix such as municipal wastewater, leading to optimized process design and control.

Using renewable energy to power chemical oxidation systems can help reduce operating costs and the associated environmental impact of energy. Solar-induced energy harvesters coupled with photocatalytic AOPs have shown potential for decentralized, remote treatment (Huo et al. 2021). Also, using chemical oxidation methods paired with advanced primary treatment would eliminate the need for secondary sludge handling

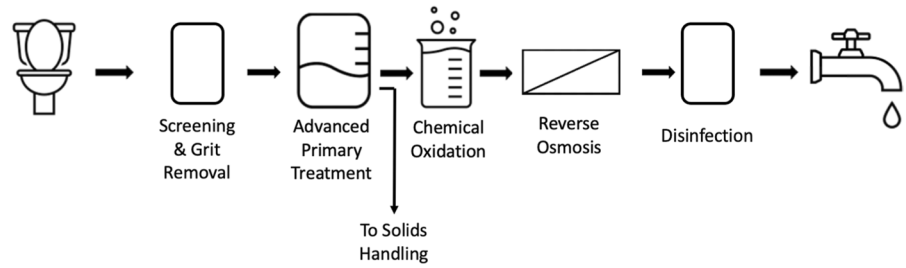


Fig. 4 Proposed integrated DPR treatment system

while also increasing the amount of primary sludge available for anaerobic digestion and subsequent methane production for renewable energy. This could increase the amount of energy recovered from water treatment, reducing the overall energy demand.

6.2 Kinetic evaluation of chemical oxidation systems

Before any chemical oxidation systems could be implemented for municipal wastewater, an understanding of their kinetics is needed. Studies have been conducted to determine kinetics for ozone and AOP chemical oxidation, but focused on tertiary treatment (i.e., oxidation of secondary effluent wastewater); no reports were found that describe chemical oxidation kinetics specifically for secondary treatment of municipal wastewater (Zimmermann et al. 2011; Lee et al. 2013; Thalmann et al. 2018; Lee and Gunten 2010). Without this knowledge, there is no way to determine how much chemical or what hydraulic retention time is needed. Additionally, there is no information on how these systems will behave differently with differing temperatures or differing municipal wastewater matrices.

6.3 Nutrient removal in chemical oxidation systems

One main advantage of biological treatment compared to chemical oxidation is the ability to remove nitrogen and phosphorus. Chemical oxidation does not directly remove nutrients, but it can convert nutrients to more reactive forms for subsequent removal by other processes. Additional research is needed to determine what chemical oxidation method is the most effective at converting nutrients to a more readily removable form, what the rates of conversion are, and what process should follow to remove and recover nutrients.

7 Conclusions

Secondary biological treatment such as activated sludge has been a critical component in many WRRFs since the late 1800s. Biological treatment is advantageous as it can effectively remove BOD, with some systems able to remove nitrogen and phosphorus as well. Yet, biological treatment has challenges as it cannot effectively remove CECs and ARB/ARGs, has many operational issues (e.g., sludge bulking, toxicants, and biomass washout), generates WAS that must be handled, has long detention times (and thus large footprints), and requires energy for aeration.

Chemical oxidation systems offer a potential alternative to conventional secondary biological treatment, as they can effectively remove CECs and ARB/ARGs, integrate disinfection, have shorter HRTs (and thus smaller footprints), do not generate WAS, and avoid many operational issues associated with biological treatment. These advantages make secondary chemical treatment potentially applicable to advanced wastewater treatment, decentralized treatment, wet-weather/high-flow treatment, and potable water reuse.

Before chemical oxidation can be more fully considered as an alternative to biological processes for secondary treatment, future research is needed to understand or improve energy efficiency, kinetics of sewage chemical oxidation, possible toxic byproducts and DBPs, nutrient removal, and integration with other treatment processes, especially to achieve nutrient removal/reuse and more sustainable solids management.

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