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Review Article

Ozonation Processes for the Removal of Acetaminophen from Wastewater: A Systematic and Bibliometric Review

Procesos de ozonización para la eliminación de acetaminofén en aguas residuales: revisión sistemática y bibliométrica

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Abstract

Introduction: Acetaminophen is an emerging contaminant of growing concern due to its presence and persistence in water bodies. Conventional wastewater treatment processes have limitations in removing pharmaceutical compounds and may achieve low levels of mineralization, favoring the formation of potentially toxic intermediate by-products. In this context, catalytic ozonation emerges as an advanced technology to improve their degradation and mineralization.
Objective: To analyze the behavior and efficiency of ozonation and catalytic ozonation processes for the removal of acetaminophen from wastewater.
Methodology: A total of 82 scientific articles published between 1988 and 2025 were collected through a systematic search in Scopus. The information was processed using VOSviewer to characterize research trends and structure a specialized technical review.
Results: The bibliometric results showed a sustained increase in scientific interest since 2013, with a peak between 2017 and 2022, mainly associated with ozonation, pharmaceuticals, and wastewater treatment. The technical review confirmed the limitations of conventional biological treatments for recalcitrant compounds such as acetaminophen. In contrast, catalytic ozonation, particularly using catalysts such as magnetite, showed greater degradation and mineralization capacity, reaching up to 99.2%, compared with approximately 51% COD reduction reported for simple ozonation.
Conclusions: Catalytic ozonation is positioned as a promising alternative for the advanced treatment of wastewater containing pharmaceutical contaminants.

Keywords: Emerging contaminants, Hospital effluents, Catalytic ozonation, Paracetamol, Advanced oxidation processes, and hydroxyl radicals.

Resumen

Introducción: El acetaminofén es un contaminante emergente de creciente preocupación por su presencia y persistencia en cuerpos de agua. Los procesos convencionales de tratamiento de aguas residuales presentan limitaciones para eliminar compuestos farmacéuticos y pueden alcanzar bajos niveles de mineralización, favoreciendo la formación de subproductos intermedios potencialmente tóxicos. En este contexto, la ozonización catalítica se perfila como una tecnología avanzada para mejorar su degradación y mineralización.
Objetivo: Analizar el comportamiento y la eficiencia de los procesos de ozonización y ozonización catalítica para la remoción de acetaminofén en aguas residuales.
Metodología: Se recopilaron 82 artículos científicos publicados entre 1988 y 2025, seleccionados mediante una búsqueda sistemática en Scopus. La información se procesó mediante VOSviewer para caracterizar las tendencias de investigación y estructurar una revisión técnica especializada.
Resultados: Los resultados bibliométricos evidenciaron un incremento sostenido del interés científico desde 2013, con un pico entre 2017 y 2022, asociado principalmente con ozonation, pharmaceuticals y wastewater treatment. La revisión técnica confirmó las limitaciones de los tratamientos biológicos convencionales frente a compuestos recalcitrantes como el acetaminofén. La ozonización catalítica, particularmente mediante catalizadores como la magnetita, mostró mayor capacidad de degradación y mineralización, alcanzando hasta 99,2%, frente a aproximadamente 51% de reducción de DQO reportada para la ozonización simple.
Conclusiones: La ozonización catalítica se posiciona como una alternativa prometedora para el tratamiento avanzado de aguas residuales con contaminantes farmacéuticos.

Palabras clave: Contaminantes emergentes, Efluentes hospitalarios, Ozonización catalítica, Paracetamol, Procesos de oxidación avanzada y radicales hidroxilo.

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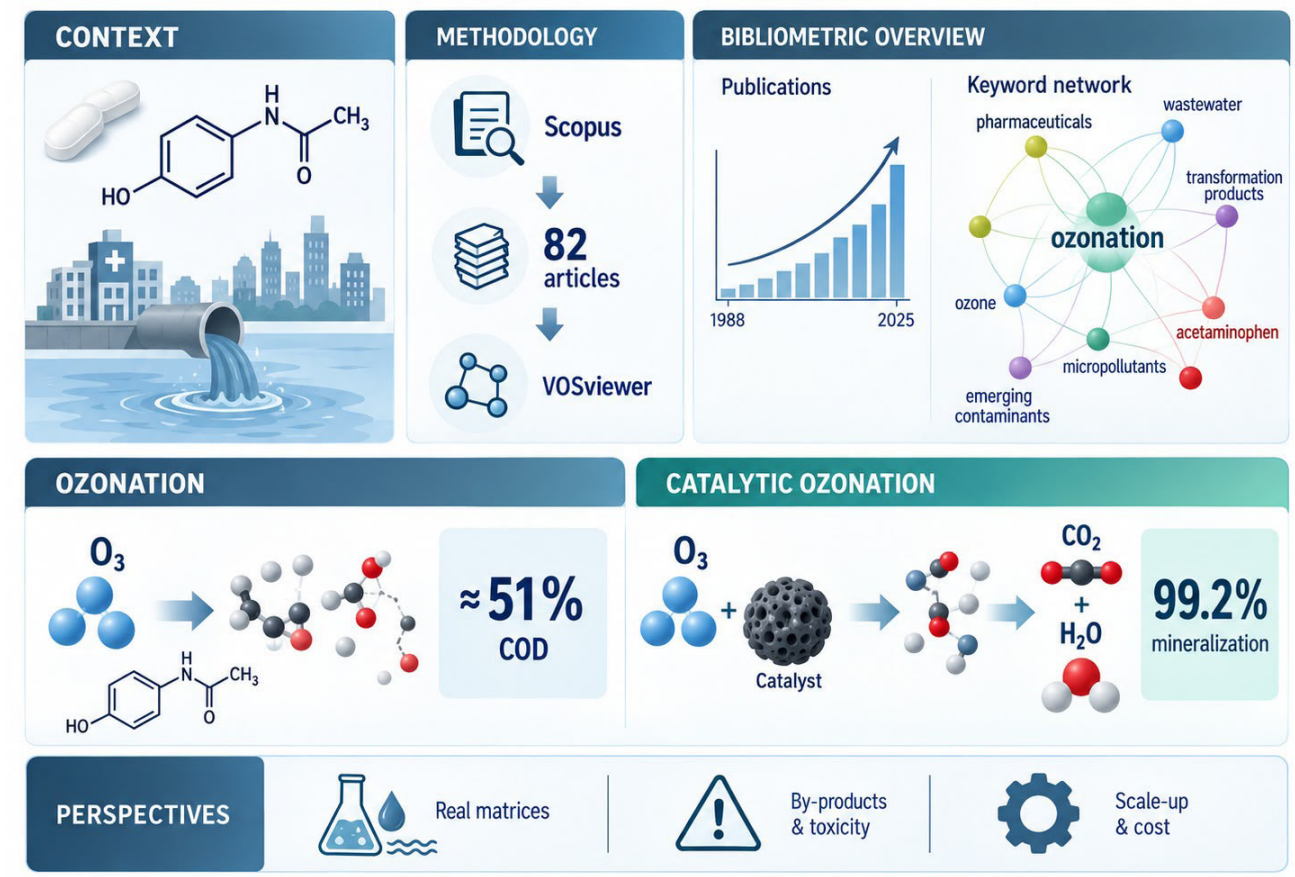
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Why was the study conducted?
To systematically assess the application of ozonation-based advanced oxidation processes for acetaminophen removal from wastewater, addressing the limitations of conventional treatment and identifying research gaps related to mineralization, transformation products, toxicity, and real-world application.

What were the key findings?
The review showed that catalytic ozonation generally provides higher degradation and mineralization efficiencies than conventional ozonation, with some catalytic systems achieving up to 99.2% mineralization. However, performance depends strongly on operating conditions and wastewater matrix, while the toxicity of transformation products and large-scale application remain insufficiently evaluated.

What do these findings contribute?
The findings provide a structured overview of the scientific development and technical performance of ozonation-based technologies for acetaminophen removal, while highlighting priorities for future research, including real wastewater validation, catalyst reuse and economic viability, ecotoxicological assessment, and life-cycle analysis.

Graphical Abstract



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Introduction

Growing global concern about water quality has intensified due to the presence of emerging contaminants (ECs). ECs are substances, such as pharmaceuticals and personal care products, found in the environment that can have adverse effects on ecosystems and human health(1,2)**.** Emerging contaminants have gained increasing significance in recent decades due to their frequent detection in surface water bodies, groundwater, and drinking water supply systems. Despite the potential risks associated with their presence, many of these compounds are not yet specifically regulated under various environmental laws. This situation has spurred research aimed at understanding their environmental behavior, assessing their ecotoxicological risks, and strengthening management and control strategies. Furthermore, there is a recognized need to implement more efficient treatment technologies that minimize the environmental, economic, and public health impacts associated with the presence of emerging contaminants in water resources(3,4).

Acetaminophen (ACT), also known as paracetamol (ACT), a pain reliever and fever reducer widely used worldwide (5), has been repeatedly detected in various aquatic matrices (6,7). These include drinking water, wastewater, surface water, groundwater, and even landfill leachate (8). Its presence is attributed to its widespread use, incomplete excretion by humans and animals, effluent discharges from the pharmaceutical industry, and the improper disposal of expired or unused medications (9). Despite variations in concentrations, only about 80–92% of this amount is degraded during conventional treatment processes, resulting in a significant discharge of ACTs into natural water bodies (8). The accumulation of ACT and its metabolites can cause severe ecotoxic effects, including damage to the genetic code, oxidative lipid degradation, protein denaturation in cells, and demonstrated toxicity in bacteria, algae, macrophytes, protozoa, and fish (10). Specifically, concentrations exceeding 1.8 µg/L of ACT can induce toxic responses in aquatic life (6).

Given this scenario, conventional wastewater treatment methods, such as biological processes (11) and filtration (5), have proven ineffective in completely removing ACT and other recalcitrant pharmaceutical compounds (12). The complexity of these drugs' molecular structure and their physicochemical properties do not facilitate their degradation by traditional biological processes(13) . Furthermore, primary treatment processes achieve removal efficiencies of only 20% to 50% for various pharmaceutical compounds(11) , and the partial oxidation of these contaminants can lead to the formation of more toxic byproducts, as has been observed with ozone, which can form N-nitrosodimethylamine (14). Although conventional wastewater treatment systems (WWTPs) can achieve apparent removal rates of up to 80–92% for acetaminophen (15), residual concentrations in effluents remain a cause for environmental concern, with values of 21.357 ng/L in hospital effluents in China, well above those recorded in previous studies in Vietnam (10.734 ng/L) and Greece (9.300 ng/L) (16). This comparison reveals a trend toward higher residual concentrations reported in more recent studies compared to research conducted in previous years, suggesting greater persistence and presence of this contaminant in aquatic systems**.** These concentrations far exceed the aquatic toxicity threshold (1.8 µg/L) (17) and are associated with chronic effects in organisms such as rainbow trout, including kidney, gill, and liver damage (7)**.** This persistence confirms the inadequacy of conventional treatment methods and underscores the need to develop more effective and sustainable technologies to prevent the ecotoxicological risks arising from the continuous discharge of emerging contaminants (11).

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Within the field of Advanced Oxidation Technologies**,** the ozonation technique and its catalytic variants have emerged as promising solutions for the removal of emerging contaminants(18) . Ozone (O_3) is a powerful oxidizing agent with a high standard reduction potential (E° = 2.07 V/SHE) (9), capable of reacting directly with organic compounds or decomposing in water to generate even more reactive oxidizing species, such as hydroxyl radicals (•OH, E° = 2.8 V/SHE) (9) . Unlike other reagents, ozone can be generated in situ, eliminating the need to transport and store hazardous materials, and it does not produce chlorinated byproducts in actual wastewater (7). However, conventional ozonation faces limitations such as incomplete mineralization and the potential formation of byproducts that are more toxic than the original compound, such as primary aromatic intermediates, secondary aromatic derivatives, aliphatic carboxylic acids, and persistent acids (oxalic and oxamic) (19). This knowledge gap—due to the scarcity of comparative studies on residual toxicity and complete mineralization in real matrices—highlights the need to refocus efforts toward strategies that optimize the generation of hydroxyl radicals (•OH) and minimize the toxicological risks associated with intermediate byproducts (20).

Ozonation processes, particularly those that incorporate catalysts or are combined with other advanced oxidation technologies (20), have demonstrated significantly improved efficiency in the degradation of recalcitrant contaminants (21). In the specific case of acetaminophen (ACT), its rapid degradation via simple ozonation has been documented (9) , and even its complete removal has been achieved in enhanced treatments using micro-nano ozone bubbles or in combination with electrolysis, reaching removal rates of 99.5%(22). Ozonation can also improve the biodegradability of the effluent, facilitating subsequent treatments (9) .

Based on the evidence reported in the scientific literature, it is proposed that catalytic ozonation and ozone-based hybrid processes exhibit greater efficiency in the degradation and mineralization of acetaminophen in wastewater compared to conventional ozonation, due to the increased generation of highly reactive oxidizing species, particularly hydroxyl radicals (•OH). In this context, the present review aims not only to synthesize scientific advances regarding the application of ozonation technologies for acetaminophen removal but also to conduct a critical analysis of their limitations, identifying research gaps related to incomplete mineralization, the formation of toxic byproducts, and the lack of studies in real-world matrices (9,23)).

Methodology

Literature Review

This study conducted a systematic literature review accompanied by a bibliometric analysis to evaluate scientific trends related to the degradation of ACT using ozone-based advanced oxidation processes. The literature search was conducted in Scopus from 1998 to 2025 to reflect the evolution of interest in pharmaceutical contaminants and advanced treatment technologies**.**

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To identify relevant studies, keywords and Boolean operators were used: ("acetaminophen" OR "paracetamol") AND ("ozonation" OR "ozone-based advanced oxidation processes" OR "O_3 AOP" OR "catalytic ozonation") AND ("wastewater treatment" OR "aqueous solution" OR "effluent"). This search strategy was specifically applied to the titles, abstracts, and keywords of the publications to refine the results and ensure that the documents were related to both the environmental relevance of the contaminant and the treatment technologies. Inclusion criteria were applied to select the most relevant literature. Only scientific articles and review articles published in English were included.

Bibliometric Analysis

The analysis focused primarily on the co-occurrence of keywords (author keywords) to explore the main concepts and topics related to the degradation of ACTs via ozonation. A minimum of 5 occurrences per keyword was established as an inclusion criterion, along with a relevance threshold of 12, which allowed for filtering the most significant terms within the analyzed corpus. This configuration helped reduce bibliographic noise and focus the analysis on the most relevant nodes of the thematic network.

In addition to thematic co-occurrence, the temporal evolution of the keywords was analyzed, allowing us to observe changes in research trends over time. To this end, VOSviewer assigned a color to each term based on the average publication year in which it appears, which made it easier to identify emerging, established, or declining topics within the field of study.

The bibliometric maps were constructed using the file exported from Scopus, which had been previously cleaned to remove duplicates and normalize equivalent terms (for example, "advanced oxidation processes" and "AOPs") (24). The generated maps show both the strength of the link between terms and their frequency of occurrence, using the full-counting method. The resulting visualization allowed us to identify thematic clusters representing subsets of publications with a high degree of semantic similarity (25).

This bibliometric analysis provided a solid quantitative basis for understanding the structure and dynamics of scientific knowledge regarding the use of ozonation for the removal of ACTs, while also serving as a basis for identifying gaps and opportunities for future research.

Results

Bibliometric analysis of research trends using VOSviewer

The bibliometric analysis allowed us to identify the evolution and structure of scientific output related to the removal of ACTs through ozonation processes, using 82 articles published between 1988 and 2025, as shown in Figure 1.

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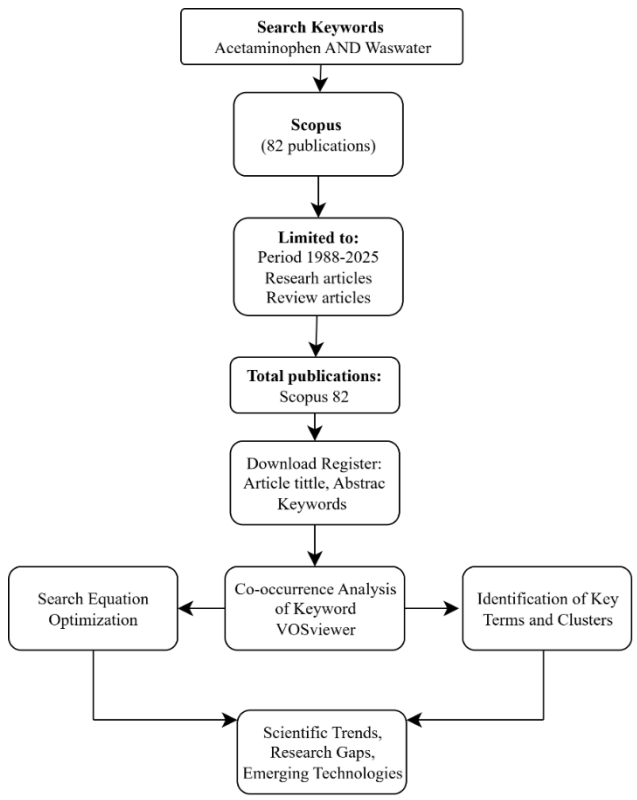


Figure 1. Selection process and bibliometric analysis in Scopus. Author's own work (2025).

The temporal evolution of publications shows a sustained increase beginning in 2013, with a more intense concentration between 2017 and 2022. This trend reflects a growing concern about the environmental impact of pharmaceutical pollutants and an interest in applying advanced technologies such as ozonation and catalytic ozonation in wastewater treatment.

Figure 2 shows the term co-occurrence map generated using VOSviewer, which revealed that the most frequent keywords in the analyzed dataset include ozonation, acetaminophen, paracetamol, pharmaceuticals, wastewater treatment, catalytic ozonation, ozone, micropollutants, water treatment, and transformation products. These words are grouped into thematic clusters that allow for the visualization of well-defined lines of research. The term "ozonation" appears as a central node, strongly connected to topics related to emerging contaminants, transformation products, and advanced oxidation technologies, suggesting its dominant role in the current literature. The density analysis confirms that this term accounts for the highest number of publications and co-occurrences, followed by "wastewater treatment" and "pharmaceuticals."

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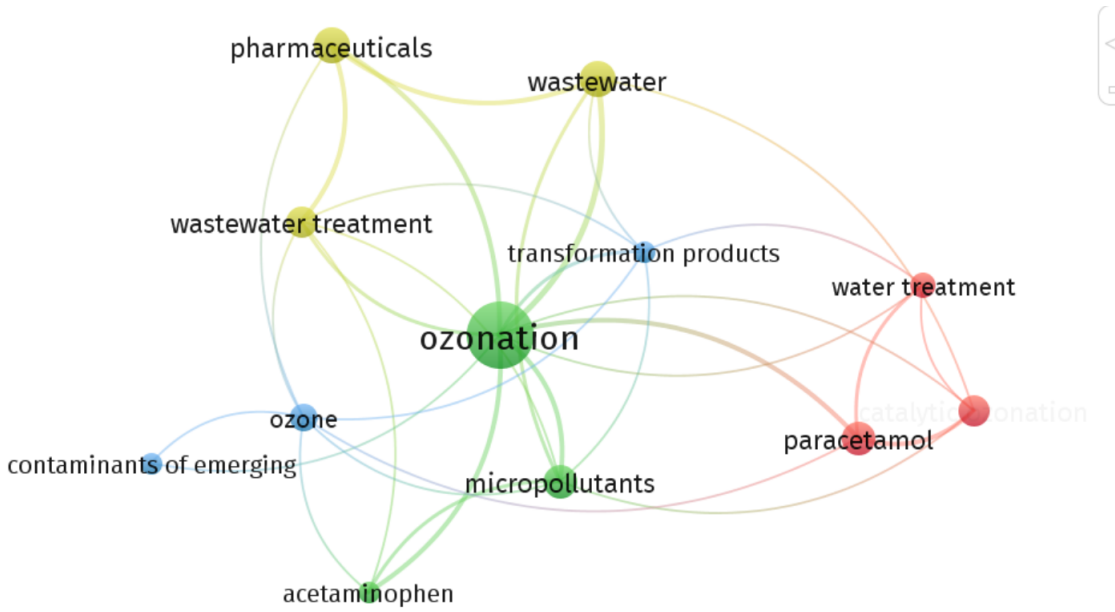


Figure 2. Co-occurrence map of keywords related to acetaminophen in wastewater. Source: Prepared by the authors using VOSviewer with data exported from Scopus (1988–2025).

At the level of authors, institutions, and countries, the results show a significant concentration of research in European and Asian countries, with notable contributions from China, Spain, Iran, and Germany. Co-authorship networks suggest the existence of established research groups, with inter-institutional collaborations that have strengthened the development of experimental studies on the degradation of acetaminophen and other pharmaceutical substances. Publications linked to universities with active research lines in advanced oxidation processes reflect the field's growing maturity and its specialization toward sustainable treatments.

Figure 3, which shows the temporal evolution, indicates that concepts such as paracetamol, ozonation, and micropollutants began to gain prominence in the literature around 2015, while the term "catalytic ozonation" shows more recent growth, establishing itself as an emerging area of research. This suggests a transition from the study of conventional processes toward an interest in more specific and reactive technologies. However, gaps have also been identified in comparative studies between technologies, the evaluation of intermediate products, and comprehensive ecotoxicological analyses, reinforcing the need for future experimental research aimed at optimizing processes such as catalytic ozonation using materials like magnetite**.**

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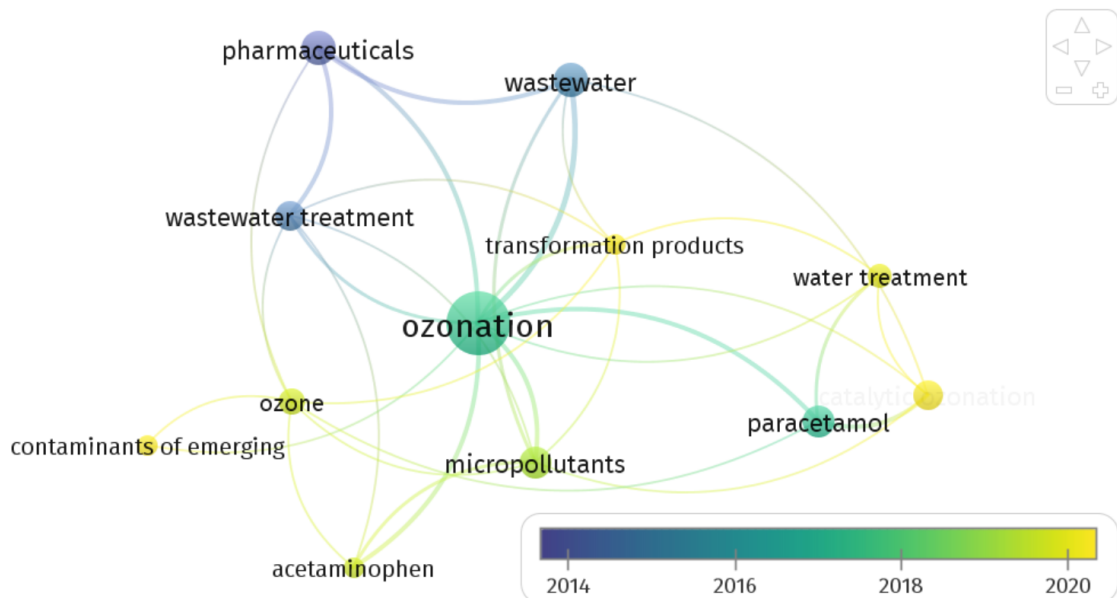


Figure 3. Temporal network of co-occurrences of keywords related to acetaminophen in wastewater. Source: Prepared by the authors using VOSviewer with data exported from Scopus (1988–2025).

Acetaminophen in wastewater

ACT is an analgesic and antipyretic drug widely used around the world. Its global annual intake is estimated at nearly 1.45 metric tons per year. However, it should be noted that this estimate is based on data reported in 2020 and may have changed in subsequent years (26)). After consumption by humans and animals, 90% of ACT is excreted unmetabolized or in conjugated forms through urine, entering sewer systems and subsequently wastewater treatment plants (WWTPs) and other natural aquatic environments (10,26). In addition to excretion, discharges from pharmaceutical industries and the disposal of expired or unused medications also contribute significantly to their presence in the aquatic environment (8,26)). Hospital effluents are a major source of drug conjugates, antibiotic resistance genes, and pharmaceutical metabolites (27). The annual inflow of ACTs to wastewater treatment facilities is estimated to be between 292 and 585 metric tons globally, of which only approximately 80% is degraded during treatment, resulting in the discharge of large quantities into natural waters (8).The presence of pharmaceuticals and their metabolites poses a risk to global health due to their persistence, bioaccumulation, and biomagnification, which can alter the behavior and reproductive health of aquatic organisms, promote the spread of antibiotic resistance, and cause long-term ecological imbalances (28).

The presence of ACTs in the aquatic environment is a cause for concern due to their documented ecotoxicological effects. It has been demonstrated that ACTs can cause damage to the genetic code, oxidative degradation of lipids, and denaturation of proteins in cells, with well-established toxicity in bacteria, algae, macrophytes, protozoa, and fish (9,10). Specifically, non-narcotic analgesics such as acetaminophen can induce oxidative stress in cyanobacteria, reducing chlorophyll and carotenoid content and inhibiting growth (11). Although it exhibits low toxicity to Daphnia (with

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lethal and reproductive effects in the mg/L range), acetaminophen can induce oxidative stress and reproductive dysfunction, especially in crustaceans (16). Concentrations of ACT exceeding 1.8 µg/L can cause toxic responses in aquatic life (8)), and it has been observed that its degradation byproducts—such as Primary Aromatic Intermediates, Secondary Aromatic Derivatives, Aliphatic Carboxylic Acids, and Persistent Acids (oxalic and oxamic)—may be significantly less harmful than the original compound (29).

ACT is frequently detected in various aquatic matrices, including wastewater, surface water, and groundwater (26). Concentrations in the influents of municipal wastewater treatment plants (WWTPs) range from not detected (ND) to 66,440,000 ng/L, while ranges from ND to 90,500 ng/L have been reported in effluents (30). The removal of acetaminophen in conventional municipal WWTPs ranges from 14.3% to 100%, with an average efficiency of 94.7% (30). Despite these high removal efficiencies, levels of ACT have been found in WWTP effluents as high as 10 mg/L (17).

A study in Spain found that acetaminophen was highly removed (greater than 70%) at the three WWTPs investigated. At WWTPs with advanced treatment, the total concentration of pharmaceuticals (diclofenac, losartan, tramadol, O-desmethyl-venlafaxine, chlorpyrifos, and terbutryn), which was approximately 263 µg/L in the influent, was reduced to 14 µg/L in the effluent, demonstrating an efficiency of over 90% for ACT (31). However, removal in raw hospital effluents shows significant variability, ranging from 75.56% for lidocaine to 100% for other compounds, with overall removal efficiencies ranging from 10.23% to 64.01% (14). In the case of acetaminophen, it was detected in 100% of the hospital influent samples and in 67% of the effluent samples, demonstrating its persistence in hospital wastewater and the need to implement more efficient treatment processes for its removal (16). Conventional activated sludge systems face challenges in removing hydrophilic compounds such as acetaminophen, caffeine, metformin, and certain antibiotics, often requiring advanced oxidative treatments (16).

Emerging risks associated with the persistence and toxicity of ACT and its transformation products underscore the need for adequate pretreatment prior to discharge to preserve environmental quality and mitigate its harmful effects (26). During the degradation of acetaminophen via advanced oxidation processes based on ozonation, transformation by byproducts such as p-aminophenol, hydroquinone, benzoquinones, and various low-molecular-weight organic acids may be generated (32). Some of these compounds may exhibit greater persistence or toxicity than the original contaminant, making their identification and evaluation essential (32). However, the available information on the toxicity of several of these intermediates is still limited, highlighting the need to develop methodologies to monitor the formation and evolution of the products generated during treatment (23).

Advanced oxidation processes based on ozonation

Ozonation, which uses ozone (O_3), has established itself as an effective advanced oxidation process for the treatment of industrial wastewater (33). Ozone, an unstable and toxic gas, rapidly converts to oxygen ($_{O_2}$) and must be continuously generated on-site using an ozonator (9). This powerful oxidizing agent, with a standard reduction potential (E°) of 2.07 V/SHE, is capable of breaking down a wide range of organic and inorganic pollutants (21,33). Its oxidative action is based on two main

doi: 10.25100/iyc.v28i2.15651

mechanisms: direct oxidation by O_3 molecules and indirect oxidation via reactive oxygen species (ROS), particularly hydroxyl radicals (•OH) generated by the decomposition of ozone (21,34). The •OH, with a reduction potential of 2.8 V/SHE, is the most potent oxidizing agent known in ozonation systems (35,36). In simplified terms, this process can be represented by the following reactions:

$$O_3 + OH^- \rightarrow HO_2^- + O_2 \quad (1)$$

$$HO_2^- \rightleftharpoons O_2\bullet^- + H^+ \quad (2)$$

$$O_3 + O_2\bullet^- \rightarrow O_3\bullet^- + O_2 \quad (3)$$

$$O_3\bullet^- + H^+ \rightarrow HO_3\bullet \quad (4)$$

$$HO_3\bullet \rightarrow \bullet OH + O_2 \quad (5)$$

In ozonation systems that use ozone alone, the predominant oxidation of TOC and its byproducts occurs via $_{O_3}$, with the participation of the •OH radicals generated (9). However, the low autolysis rate of ozone limits the sufficient production of •OH, which restricts the removal of hard-to-degrade contaminants and can lead to the formation of carboxylic acid intermediates, reducing the mineralization rate (21)). For example, in one study, conventional ozonation of effluents treated with 3-chlorophenol and chlorobenzene showed a reduction in effluent toxicity ranging from 41% to 83% during the first four days of storage at 22 °C (37). Although simple ozonation is effective for the degradation of acetaminophen, the mineralization of the organic compounds present is often limited. In this regard, a 51% reduction in COD has been reported after 120 minutes of treatment, indicating partial mineralization and the persistence of transformation byproducts, such as oxalic acid (9) . However, direct ozonation proved effective for the oxidation of most drugs (>90%) in controlled studies, although the removal of ibuprofen was only possible by combining O_3 with H_2O_2 (38).

The combination of ozone and hydrogen peroxide (O_3/ H_2O_2), known as the peroxono process, is a promising advanced oxidation process (AOP) that significantly enhances the generation of hydroxyl radicals (33,39). This addition increases the process's efficiency, since one molecule of $_{H_2O_2}$can produce one •OH radical, in contrast to the three ozone molecules required to achieve the same effect in conventional ozonation (40). The O_3/H_2O_2 process achieved degradation efficiencies of 92% for ketoprofen (KET), 86% for diclofenac (DCF), and 78% for their mixture, with a notable improvement in the physicochemical quality of the effluent, including reductions in turbidity, conductivity, COD, Biochemical Oxygen Demand (BOD_5) and Total Organic Carbon (TOC) (38). In addition, a substantial reduction in toxicity was observed. The presence of chloride (Cl-) and bromide (Br-) ions in the solution can, in some cases, accelerate the degradation of TOC in the UV/ H_2O_2 process, due to the substantial generation of reactive halogen species (41).

Catalytic ozonation processes represent a further improvement over conventional ozone treatment by enhancing the conversion of O_3 to •OH (21). Heterogeneous catalytic ozonation facilitates easy separation of the treated water, which contributes to its economic viability (21). Metal oxide-

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based catalysts, such as ZnO, CeO_2, γ-Al_2O_3, and MnO_2–CeO_2, have demonstrated excellent catalytic potential(20). The dispersion of $_{CeO_2}$ nanoparticles in diatomite (CeO_2@DE) significantly increased the degradation of ciprofloxacin (CIP) from 47.2% to 94.2% and its mineralization to 76.8% (20). For ACT, ozonation using an Fe_3O_4@Ce-UiO-66 catalyst achieved 90.2% mineralization (42). The magnetic carbon biomaterial (Fe-N@BC) activated ozone for the degradation of tetracycline (TC) with an efficiency of up to 95.0% and minimal interference from coexisting anions (42). Catalytic ozonation using modified MgO nanoparticles demonstrated an 87% reduction in ACT (9).

The efficiency of ozonation is strongly influenced by factors such as pH, temperature, and the composition of the water matrix (43). ACT degradation is faster at higher pH levels (e.g., pH 10.0 compared to pH 2.0 or neutral pH), due to faster oxidation of the phenolate form of ACT—which is more reactive—and increased production of •OH (9). For the degradation of phenol via UV/H_2O_2, increasing the pH from 3 to 8 attenuated the inhibitory effects and reduced the acute toxicity of halogenated byproducts (41). As for temperature, it significantly affects the rate and extent of biotoxicity attenuation in ozonated effluents, with higher temperatures facilitating a faster and more complete attenuation of toxicity (37). The presence of complex matrices, such as actual wastewater, significantly affects the efficiency of ozonation (15,44). Dissolved organic matter (DOM) competes for dissolved ozone and hydroxyl radicals, reducing the concentration of oxidant available for the contaminants of interest (44). Therefore, in hospital wastewater with high DOM concentrations (up to 500 mg/L), direct ozonation may be economically unfeasible; consequently, it is often used as a polishing step following biological treatments, where the DOM concentration is lower (6–8 mg/L) (44). However, the applicability of simple ozonation for the remediation of multicomponent activated sludge systems and hospital wastewater on a full-scale basis has been demonstrated (9).

Compared to other AOPs, ozonation offers distinct advantages. It does not require the transport or storage of hazardous materials, since ozone is generated on-site (9). It has a greater oxidation capacity than active chlorine and other conventional methods, and does not produce chlorinated organic byproducts in actual wastewater, eliminating the need for dichlorination (9). Furthermore, ozone has a half-life of 20 minutes in water, readily decomposing into O_2, which prevents residual toxicity after treatment (9). In wastewater treatment plants, ozonation has demonstrated greater efficiency in COD removal (9). A comparative study of six different AOPs for micropollutants showed that ozonation had the lowest energy consumption among the processes evaluated (43). Another study indicated that ozonation had the highest energy efficiency among five AOPs investigated (15). However, a comparative study found that direct ozonation was not efficient for the degradation of acetaminophen, and the addition of H_2O_2 did not result in a significant improvement, whereas heterogeneous photocatalysis showed better performance (64% removal) (45). This highlights the importance of considering specific conditions and the properties of the contaminants.

Mechanisms of acetaminophen degradation by ozonation

The degradation of ACT by ozonation occurs through direct oxidation by (O_3) molecules and indirect oxidation (21). The indirect pathway via •OH is particularly effective in enhancing the mineralization of contaminants(21). Studies have shown that, regardless of the ozonation process used, similar byproducts are detected, suggesting that the same oxidizing agents (O_3 and •OH) act on organic contaminants (9). The degradation of ACT, in particular, can follow ionic or radical

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mechanisms; in the ionic process, O_3 directly attacks the aromatic chain, whereas in the radical process, HO• and/or O_2 radicals contribute to the formation of similar products, with both pathways competing (40). It has been observed that the drug's disappearance rate is faster at higher pH levels, which is attributed to the faster oxidation of the phenolate form of ACT (more reactive at pH 10.0) and to greater production of •OH radicals, which react more rapidly with ACT than ozone itself (9). Although direct ozonation is the predominant process in the oxidation of ACT and its byproducts, involving the generated •OH radicals, the degree of mineralization achieved tends to be low (9).

As for intermediates and end products, the ozonation of ACT produces hydroquinone and 2-hydroxy-4-(N-acetyl)aminophenol, which in turn can generate oxalic and oxamic acids (9). Oxalic acid, in particular, has been shown to be a persistent byproduct due to its resistance to oxidation by •OH radicals (9). A common feature in the proposed reaction sequences for ACT is the initial loss of the acetamide group from ACT, leading to the formation of various aromatic compounds through oxidation of its phenolic moiety, or the direct hydroxylation of ACT (9). Subsequent breaking of the benzene ring gives rise to aliphatic byproducts that evolve into short-chain carboxylic acids, ultimately transforming into CO_2 and H_2O (9). The acetamide group also oxidizes to CO_2, releasing NO_3^- and NH_4^+ ions (9). It has been found that p-benzoquinone byproducts dominate changes in fluorescence and toxicity, and their transformation via hydroxylation and reduction leads to a decrease in the toxicity of the resulting products (37). In the case of acetaminophen, ozonation generates various intermediate transformation products whose toxicity may differ from that of the original compound. Although some studies have reported the formation of potentially more toxic byproducts during the ozonation of aromatic compounds, the available evidence for acetaminophen does not allow for generalizing this behavior; therefore, it is necessary to evaluate the toxicity of the intermediates and final products generated during treatment (37,46).

The reported mineralization rates from ozonation tend to be low, despite the high removal rates of the drugs (46). For example, for an initial ACT concentration of 50 mg/L, reductions in Chemical Oxygen Demand (COD) of 35%, 39%, and 65% were observed at pH 2.0, natural pH, and 10.0, respectively, after 60 minutes of treatment (9). One study found that, although complete degradation of acetaminophen was achieved in 20 minutes, the COD reduction was only 51% after 120 minutes (9). A kinetic analysis revealed that molecular degradation is faster than mineralization, confirming the formation of organic intermediate compounds (29).

With regard to post-treatment toxicity assessments, it is crucial to consider that, even if effective degradation of micropollutants is achieved, the formation of byproducts can lead to an increase in adverse effects such as toxicity and estrogenic activity (47) This increase in toxicity is attributed to the formation of intermediate transformation products of acetaminophen, such as p-aminophenol, hydroquinone, and p-benzoquinone. P-benzoquinone exhibits high chemical reactivity and the ability to induce oxidative stress in aquatic organisms, and may exhibit toxicity greater than that of the original compound. Likewise, ozonation without the addition of hydrogen peroxide has been shown to increase the toxicity of the effluent (38). However, it has been observed that the biotoxicity of ozonized effluents gradually decreases during storage, with reductions ranging from 41% to 83% in the first four days at 22°C, and that temperature—but not natural light—influences this attenuation of toxicity (37). In contrast, ozonated photolysis with Fe^{3+} showed an

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increase in ecotoxicity toward Daphnia magna due to the presence of this toxic ion (9). A combined advanced oxidation process (O_3 and electro-oxidation) has been shown to reduce toxicity (TCM range from 48% to 100%) and environmental risks (negligible for the original compounds, low for one compound) after 4 hours of treatment (22). Furthermore, it has been reported that the transformation products of a hybrid membrane and O_3/UV process are environmentally benign and pose no significant health risks (48).

Catalytic and combined ozonation systems

Catalytic ozonation and combined ozonation systems have demonstrated improved efficacy in the degradation of ACT and other pharmaceutical contaminants in wastewater, outperforming direct ozonation in several respects.

Heterogeneous Catalytic Ozonation

Various catalytic materials have been investigated. For example, a biochar nanocomposite internally doped with Fe-N (Fe-N@BC), synthesized from iron sludge and water hyacinth, achieved a 95.0% tetracycline (TC) removal efficiency through catalytic ozonization (42). In the case of acetaminophen, mineralization reached 99.2% through ozonation using a Fe_3O_4@Ce-UiO-66 catalyst in a 1:1 ratio (49). Another study used SnO_2 nanoparticles as a catalyst, increasing ACT degradation from 84% (with simple ozonation) to 98% in 20–30 minutes, and mineralization from 34% to 83% (9). The main oxidizing agent identified was the •OH radical. An Fe_3O_4@Ce-MOF catalyst for ozone increased ACT reduction to 87% (compared to 38% with ozone alone) at pH 5.6, resulting in 55% TOC removal compared to 14% with simple ozonation (9). Iron-doped Moringa oleifera seed shells (Fe-MOSH) have shown a significantly higher ACT removal capacity (Qmax of 17.48 mg/g at 44°C) than other conventional adsorbents in catalytic ozonation (34). Furthermore, a trimetallic nMZVCC nanocomposite demonstrated enhanced heterogeneous Fenton-type catalytic activity for ACT degradation, achieving 95% removal in 180 minutes(50). A study applying catalytic ozonation to pharmaceutical wastewater observed a COD concentration in the effluent below 50 mg/L (53–59% removal efficiency), meeting the Chinese discharge standard and demonstrating massive destruction of aromatic pollutants (51).

Combined Ozonation Systems (O_3/$H_2O_2$and O_3/UV)

The combination of ozone and hydrogen peroxide (O_3/H_2O_2) has achieved degradation efficiencies of 92% for KET, 86% for DCF, and 78% for the KET+DCF mixture, with significant reductions in turbidity, conductivity, COD, and BOD_5 (38). UV irradiation combined with H_2O_2 has been shown to remove between 80% and 90% of DCF and 80% of ibuprofen within 60 minutes of irradiation(47). A hybrid system combining ultrafiltration (UF) membranes with an O_3/UV process has shown great promise for the removal of pharmaceuticals and personal care products from real wastewater (48). This system, with UF pretreatment, achieved favorable degradation of acetaminophen—80% in untreated water and 95% in biologically treated water—after 7 minutes of exposure to UV_{254} nm/1.5 $mg{\cdot}L^{-1}$of O_3 (48). However, ozonation alone, without the addition of hydrogen peroxide, has shown limited efficiency and an increase in effluent toxicity (38).

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Comparisons of Efficiency, Kinetics, and Synergy

In the comparison of AOPs, ozonation, photocatalysis, and pulsed corona discharge (PCD) in the gas phase achieved removal rates of over 90% for all detected pharmaceuticals (15). The pulsed corona discharge process showed the best energy efficiency (0.28 $kWh \cdot m^{-3}$), followed by ozonation (0.55 $kWh \cdot m^{-3}$), while photocatalysis required significantly more energy (47 $kWh \cdot m^{-3}$) (15). Although heterogeneous photocatalysis showed better ACT removal (64%) compared to direct ozonation or electro-peroxono (45), other studies indicate that electro-peroxono and ozonation showed better results for ACT degradation under the conditions investigated (45). The electro-peroxono process has proven to be more effective than simple ozonation (9). Catalytic ozone processes enhance the degradation of pollutants by promoting the conversion of O_3 to •OH radicals (21). Unlike ozonation, ozonation without the addition of $_{H_2O_2}$at Site A was found to result in poor removal of AhR activity (−84%) and estrogenic activity (29%), attributed to the mixing of reverse osmosis brine with the treated effluent (51)). Other treatment sites (B, C, and D) achieved removals of 50–91% for AhR activity and 88–95% for estrogenic activity, which is higher than previous observations for secondary WWTPs and is likely due to tertiary treatment processes (51). The biotoxicity of ozonated effluents gradually decreased during storage, with temperature—rather than natural light—being a factor influencing the attenuation of toxicity (37). It is important to note that photocatalysis alone exhibits relatively low degradation efficiency (52). A life cycle assessment (LCA) for acetaminophen removal indicated that UV-C irradiation was more sustainable than the UV-C/H_2O_2 and UV-C-driven photo-Fenton processes, primarily due to the absence of additional reagents (53).

Pilot or Full-Scale Applications

Simple ozonation has proven effective in removing ACT from activated sludge and hospital wastewater in a pilot plant, with a low energy consumption of 19 MWh/year (9). In a sequential treatment plant—which included activated sludge, membrane filtration, granular activated carbon adsorption, and ozonation—ACT was not detected after the membrane filtration step (9). Similarly, in a pilot-scale hospital wastewater treatment plant, all ACT was removed after the membrane bioreactor (MBR) process, and subsequent ozonation reduced other pharmaceuticals by more than 90% (9). Approximately 50% of the reviewed articles on ozonation focus on the treatment of actual wastewater, highlighting the practical application of this technology (9). Ozonation has been used as a key step in many wastewater treatment plants (WWTPs) (9). Injecting ozone into the influent of conventional activated sludge (CAS) systems or into the mixed liquor recirculation loop significantly improved the removal of pharmaceuticals, increasing the overall reduction from 49% (CAS alone) to 71–80% (with ozone) (44).

Challenges and Gaps in Research

Despite their advantages and proven effectiveness, ozone-based AOPs face technological limitations, particularly regarding ozone transfer in water due to its low solubility (9). High operating costs are a significant disadvantage of AOPs compared to conventional biological treatments (33). Ozone generation itself is a process that requires expensive equipment (9). Furthermore, the use of ozone in combination with other AOPs, such as O_3/H_2O_2 /UV systems, can increase electricity demand (15). Catalytic ozonation systems, although promising, often require complex and costly

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catalyst synthesis processes, which hinders their large-scale industrial application (21). Although innovative technologies such as USONiQ ozonation have been developed—which integrates ultrasound to enhance ozone transfer and hydroxyl radical formation, achieving 80% removal of micropollutants with a specific ozone dose reduced by 20% to 70% compared to standard systems—a comprehensive economic evaluation still requires further research (54).

There is a notable lack of studies conducted on a full-scale basis or using complex wastewater matrices. Most research on AOPs has focused on synthetic solutions in the laboratory, with only a limited number of studies (9 out of 91 for Fenton-type processes) addressing actual wastewater (10). This underscores the critical need for further research in complex, full-scale environments to validate industrial applicability (55,56). The effectiveness of catalysts, for example, can be inhibited in complex matrices due to competition from natural organic matter for EROs (21). Furthermore, quantifying the relative contributions of ozone and hydroxyl radicals in large-scale applications remains a challenge due to the impracticality of adding radical scavengers for regulatory and safety reasons (54).

Another significant gap is the scarcity of comprehensive studies on the toxicity of degradation byproducts. The partial oxidation of organic pollutants can, paradoxically, lead to the formation of byproducts that are more toxic than the original compounds (37,47). Although ozonation is effective in degrading pollutants, it can generate harmful intermediates that pose risks to safety and the environment (57). For example, the ozonation of certain pharmaceuticals can produce byproducts that pose greater toxicological risks (32). In the case of diclofenac and ketoprofen, ozonation without the addition of hydrogen peroxide actually increased the toxicity of the effluent, rendering it unsuitable for wastewater treatment (38). Although tools such as the ECOSAR program are used to predict the toxicity of byproducts (8,58), additional toxicological analyses are needed to fully assess the ecotoxicological risk of these systems and ensure that AOPs do not inadvertently introduce new contaminants (58).

Finally, the integration of life cycle assessments (LCA) or cost-benefit analyses is essential but still limited. Techno-economic studies are required to determine the feasibility and optimal treatment for the destruction of contaminants such as ACT(9). LCA assessments have shown that differences in environmental impact among different AOPs for ACT degradation are often negligible, and the greatest impacts are attributed to sample transport and preparation rather than to the treatment process itself (9). This highlights the importance of considering the full life cycle of these technologies to optimize processes not only in terms of removal efficiency but also in terms of long-term environmental and economic sustainability (53). The need to develop economically viable, recyclable, and durable catalytic materials is imperative for the widespread practical application of catalytic ozonation (21).

Conclusions

The systematic review and bibliometric analysis revealed sustained growth in scientific interest in the removal of acetaminophen through ozonation processes, particularly since 2013. The predominant keywords and the temporal evolution of publications show a transition from studies focused on

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conventional ozonation toward more advanced approaches, such as catalytic ozonation and hybrid advanced oxidation processes.

The analyzed results confirm that ozonation is an effective method for the degradation of acetaminophen; however, catalytic ozonation performs better because it promotes the generation of •OH, which significantly increases the mineralization of the contaminant and reduces the persistence of intermediate compounds. Catalysts such as magnetite and various metal oxides have demonstrated high degradation and mineralization efficiencies, reaching values close to 99%, which are higher than those obtained through conventional ozonation.

Furthermore, it was found that the efficiency of these processes depends on operational variables such as pH, temperature, ozone dose, catalyst type, and the composition of the aqueous matrix. Alkaline conditions favor the formation of •OH, while the presence of dissolved organic matter can reduce the availability of oxidizing species and affect treatment performance.

The review also showed that hybrid systems, such as O_3/H_2O_2, O_3/UV, and electro-peroxono, exhibit synergistic effects that improve acetaminophen degradation, reduce residual toxicity, and increase the overall efficiency of the process. However, the formation of transformation products and the assessment of their toxicity remain under-studied areas.

Finally, significant knowledge gaps were identified related to the scarcity of pilot- and industrial-scale studies, the limited ecotoxicological assessment of the byproducts generated, and the lack of comprehensive analyses of economic feasibility and sustainability. Therefore, future research should focus on validating these technologies in real matrices, developing cost-effective and reusable catalysts, and incorporating life-cycle and cost-benefit assessments to facilitate the transfer of catalytic ozonation from the laboratory to real-world wastewater treatment applications.

Statement of authorship contribution credits

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