

Editorial

# Urban and Industrial Wastewater Disinfection and Decontamination by Advanced Oxidation Processes (AOPs): Current Issues and Future Trends

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Without any doubt, the 21st century has kick-started a great evolution in all aspects of our everyday life. The tremendous advances in medicine, production of goods (such as chemicals, plastics), etc. have all contributed to an increase in the standards and the comforts of the modern society. However, combined with the growing demands in water due to the uneven population increase and product manufacturing needs, the rapid change has impacted the wastewater flows and composition of both urban and industrial origin. Environmental engineers have been facing a constantly changing landscape, where new problems seem to emerge in an alarming, “whack-a-mole”-like situation. From antibiotic resistance to microplastics, from the ever-growing list of contaminants of emerging concern to their transformation products, the need for proper, advanced wastewater treatment methods are dominating environmental engineering research to alleviate the shift of urgent needs of the water sector. Nonetheless, the multidisciplinary nature of this huge environmental problem spans a number of research topics, namely environmental, chemical and process engineering, water and sanitary engineering, environmental science, chemistry, biology and related disciplines, material science, toxicology, risk assessment, economy, social sciences, ecology and environmental policy, among others. Therefore, in order to develop efficient solution tools to tackle the urban and industrial wastewater challenges, it is of utmost importance to promote the knowledge exchange between all expertise fields involved, thus promoting a scientific, technological, and societal cooperation for a more sustainable environment and society.

It has also become clear over the past two decades that the perception of water as an infinite resource is distorted and a dangerous idea to hold on to. As a result, novel and more holistic approaches, such as effluents' reuse, the circular economy of water, the one-health integration in wastewater treatment, have gradually replaced the archaic views on treatment and discharge of wastewater. The (well-deserved) price paid for adopting such viewpoints into policy and legislation has been partially transferred to the end of the line, namely wastewater treatment plants (WWTPs). Lower limits, recovery schemes, safer disposal and handling of wastes illustrate the reality of the treatment schemes nowadays.

In 2020, the Special issue “Urban and industrial wastewater disinfection and decontamination by Advanced Oxidation Processes (AOPs): current issues and future trends” was launched to gather the innovations of the research community on (waste)water treatment worldwide. Following the call, several articles on various aspects of (waste)water treatment have been published, both in form of original research papers and literature

reviews. Authors affiliated with institutions and companies from Colombia to Switzerland, from Portugal to India and from Spain to Iran, have contributed by disseminating their research results within this Special Issue; a brief summary of the published papers follows:

Guerra-Rodríguez et al. [1] overviewed the different strategies that can be adopted in the water sector in the context of the circular economy. Wastewater reuse is a mature and well-established strategy in many water-scarce regions of the world, but its reuse poses associated risks to ecosystems and humans due to the presence of pathogens, contaminants of emerging concern, and microbial resistance genes. However, there are other options for applying the circular economy in the water sector that are still largely unexploited, such as the recovery of materials from aqueous streams or energy recovery; Guerra-Rodríguez et al. reviewed all these options, highlighting the challenges and opportunities in their application.

Camargo-Perea et al. [2] reviewed extensively the data on the degradation of emerging pollutants by ultrasound AOP, with a special emphasis on pharmaceuticals, providing a deep understanding on the operation fundamentals and the parameters that most affect its efficiency, namely, the ultrasonic frequency, the electrical power, the pH and temperature of the solution, the nature of the target contaminant, the constituents of the water matrix, and the type and the geometry of the sonochemical reactor. In addition to the applications that were detailed, this review discussed the future perspectives and the cost implications of ultrasound AOP as a possible candidate to tackle water pollution by emerging pollutants.

Guateque-Londoño et al. [3] studied the removal of the antihypertensive losartan as a pharmaceutical model in simulated fresh urine through the application of ultrasound and UVC/H<sub>2</sub>O<sub>2</sub>. They reported higher selectivity of the sonochemical process traduced by a higher ratio between the degradation rate constants obtained in fresh urine and distilled water for the removal of losartan than that obtained in the UVC/H<sub>2</sub>O<sub>2</sub> system. Although neither of the treatments reached the mineralization of the pollutant in distilled water, it was confirmed that the sonochemical system reduced the phytotoxicity of the sample, showing the potential of this technology on the regeneration of wastewater. Finally, the authors studied the generation of transformation products from the degradation of losartan, and the fitting of these results with theoretical methods to predict and rationalize the attack of degrading species. The computational analyses confirmed that the atoms on imidazole moiety of losartan were the most susceptible to transformation by the radical species, in agreement with the experimental results obtained.

The presence of antibiotics in wastewater effluents is a problem of emerging concern threatening ecosystems and human health. Moles et al. [4] evaluated the behavior of five antibiotics (amoxicillin, enrofloxacin, sulfadiazine, trimethoprim, and azithromycin) in the influent and effluent of different WWTPs located in the north of Spain. Moles et al. determined that azithromycin was the antibiotic with the highest mass load followed by enrofloxacin, trimethoprim, sulfadiazine, and amoxicillin. Regarding the behavior of these substances in the WWTPs, they observed that biological treatments have a significant influence on removal, especially the use of trickling filters that showed the highest degree of removal. Besides, the authors studied the performance of a TiO<sub>2</sub> photocatalytic treatment plant installed as tertiary treatment that minimized the Ti release into the environment and allowed catalyst reuse. This technology totally removed sulfadiazine, amoxicillin and azithromycin, while 80% of trimethoprim and enrofloxacin was removed.

Hospital effluents are the main emitters of pollutants of emerging concern such as pharmaceuticals and antibiotics. Rosero Parra et al. [5] described the treatment of hospital wastewater using two novel catalysts supported on graphene (SnO-GO and TiO<sub>2</sub>-GO). Both heterogeneous photocatalysts were demonstrated as alternatives for abatement of pollution in this kind of effluents, reducing chemical oxygen demand by 85%, the dissolved organic carbon by 94%, and 80% of phenols, allowing the discharge of the effluents in compliance with current legislation.

Karbasi et al. [6] studied a semiconductor photocatalytic material based on Bi and W oxide, as an alternative to the traditional TiO<sub>2</sub> photocatalytic disinfection process. Owing to its large band gap and exceptional structure (micro/nanohierarchical, flower-like mor-

phology) it absorbs more photons and further into the visible range, while it reduces the kinetic limitations associated with semiconductors. After the complete characterization of the newly synthesized material, *E. coli* bacteria were used as a model microorganism and the disinfection capabilities of  $\text{Bi}_2\text{WO}_6$  were found to match the benchmark  $\text{TiO}_2$  P25 under solar light but had superior bactericidal efficacy under visible light. This opens the possibilities of further studying this material and advance towards indoor applications, where sterility is a prerequisite and plain fluorescent lighting is provided.

The photocatalytic degradation of potassium hexacyanoferrate (III), as a model cyanocomplex of gold mining wastewaters, was studied by Arce-Sarria et al. [7] in a bench-scale compound parabolic collector (CPC) reactor assisted with a light-emitting diode (UV-A/LED) and a hydrothermally treated  $\text{TiO}_2$  P25, which showed a specific surface area 2.5-fold superior to the original catalyst, a slightly higher band gap energy, and a mesoporous structure. Although the  $\text{TiO}_2$  P25 performed better, the higher free cyanide release achieved with the modified material in this specific case may be beneficial for its reuse in the gold extraction process. This report opens new prospects for future research on the structural changes of the catalyst and their potentialities for photocatalytic applications.

Graça et al. [8] proposed a combined technology (ozonation followed by ultrafiltration) to simultaneously reduce organic contaminants and microbial load of secondary urban wastewater for its further reuse, complying with the legislation for water quality for irrigation. The estrogenic activity, cell viability and cellular metabolic activity were also monitored in order to assess the impact of the treatment on the biological effects. Although most organic microcontaminants were removed (except citalopram and isoproturon), the biological effects did not suggest the production of toxic by-products, and the reduction of the bacterial loads targeting the water quality standards for irrigation was effective, the authors highlighted that microbial regrowth was observed after storage, with the concomitant increase in the genes 16S rRNA and *intI1*.

Finally, Acosta-Herazo et al. [9] showed the main features of the PHOTOREAC tool developed, as well as the results in different simulations, thanks to the experience gathered by their research groups at Cartagena University (Cartagena de Indias, Colombia) and the Universidad del Valle (Cali, Colombia) during the last twenty years of research in heterogeneous solar photocatalysis, and on extensive literature research in photoreactor engineering. PHOTOREAC is an open-access application developed in the graphical user interface of MATLAB® that allows a user-friendly evaluation of the solar photoreactors operation. Furthermore, they showed the potential of the tool in several case studies such as the removal of dichloroacetic acid and methylene blue in different types of photoreactors, as well as the modeling of radiation in a Flat Plate Photoreactor.

Considering the above contributions and the current issues dominating literature, the recent challenges faced by WWTPs exceed their regular capacity, and the need for appropriate wastewater treatment calls for novel, sophisticated methods of decontamination prior to its discharge or reuse. Emerging threats such as antibiotic-resistant bacteria, antibiotic-resistance genes, and the contaminants of emerging concern (chemicals, microplastics) demand efficient, end-of-pipe solutions before their discharge into the environment or reclamation for reuse purposes. Under this scenario, AOPs have been procured as effective methods for wastewater disinfection and decontamination. Despite the huge amount of works developed in the last decade in this area, some research opportunities and future directions can be highlighted.

One important feature in any comparative analysis between different treatments or studies is the examination of the same endpoints for a safe effluent discharge or reuse, namely the abatement of chemical contaminants, mitigation of by-products, reduction of the toxicity of the effluent, bacterial inactivation, minimization of regrowth, control of antibiotic resistance determinants, and cost of treatment. This comparison is often difficult when different studies are focused in dissimilar endpoints, different matrices, and distinct operational conditions (typical at small-scale tests). It is important to highlight that larger-scale studies than some published in this Special Issue and a further cost effectiveness

analysis of scaled-up processes are valuable for a comprehensive analysis of any given treatment option. In this regard, solar-driven AOPs are very promising when compared with other advanced technologies, being a research topic that has plenty room for further advances, for instance by developing and tailoring new photocatalysts aimed to be active under sunlight.

Regarding the endpoints, the development of tools to predict the fate and behavior of emerging contaminants during wastewater treatment would be a great asset. Given that the elimination of chemical and biological contaminants of emerging concern from wastewater by AOPs largely depends on the composition of the water matrix, studies conducted under environmental-like conditions are crucial to advance our knowledge on AOPs. In this sense, environmental relevant concentrations of mixtures of contaminants and realistic water matrices should be used instead of purely artificial conditions because the efficiency of each process results from a complex balance between inhibitory and promoting effects originated by the water components. The available data about such matrix effects on the elimination efficiency of contaminants in WWTPs is still limited and research towards the impact of the main water/wastewater constituents and the mechanisms governing it are required. These environmental-like conditions are also imperative for a better understanding of the competition effects between different contaminants; thus, the approach of studying spiked synthetic solutions spiked with one or few compounds at concentrations orders of magnitude above than those found in the environment tends to become obsolete, except for some specific applications such as performance studies of new catalysts, elucidation of degradation pathways, and characterization of chemical structures of transformation products.

As far as the fate of contaminants during treatment is concerned, the structural elucidation of the transformation products is pivotal for an accurate evaluation of the degradation pathways and for a deep understanding of their possible side-effects, like toxicity. The ecotoxicological studies provide a useful tool to clarify the harmful effects of both parent contaminants and their transformation products on the ecosystems and human health, as well as to understand the possible impact of the reagents and catalysts used. Combining other technologies can be an interesting option to deal with toxic transformation products, for instance by coupling an adsorptive post-treatment. On the other hand, AOPs are very promising as post-treatment to treat the resulting concentrate of membrane-driven processes.

Concluding, the water/wastewater treatment is a hot topic of research that has been challenged in the last decade by newly identified threats (e.g., antibiotic resistance, microplastics) that have provided new perspectives and recent opportunities for investigation. Besides the importance of bearing in mind the multidisciplinary of this topic and the need for knowledge exchange between different subjects in order to generate innovation and scientific advances, raising awareness within society and promoting individuals' involvement in the mitigation of wastewater problems is a prerequisite for a more proficient development and implementation of possible solutions.

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## References

1. Guerra-Rodríguez, S.; Oulego, P.; Rodríguez, E.; Singh, D.N.; Rodríguez-Chueca, J. Towards the Implementation of Circular Economy in the Wastewater Sector: Challenges and Opportunities. *Water* **2020**, *12*, 1431. [[CrossRef](#)]
2. Camargo-Perea, A.L.; Rubio-Clemente, A.; Peñuela, G.A. Use of Ultrasound as an Advanced Oxidation Process for the Degradation of Emerging Pollutants in Water. *Water* **2020**, *12*, 1068. [[CrossRef](#)]
3. Guateque-Londoño, J.F.; Serna-Galvis, E.A.; Ávila-Torres, Y.; Torres-Palma, R.A. Degradation of Losartan in Fresh Urine by Sonochemical and Photochemical Advanced Oxidation Processes. *Water* **2020**, *12*, 3398. [[CrossRef](#)]
4. Moles, S.; Mosteo, R.; Gómez, J.; Szpunar, J.; Gozzo, S.; Castillo, J.R.; Ormad, M.P. Towards the Removal of Antibiotics Detected in Wastewaters in the POCTEFA Territory: Occurrence and TiO<sub>2</sub> Photocatalytic Pilot-Scale Plant Performance. *Water* **2020**, *12*, 1453. [[CrossRef](#)]
5. Rosero Parra, L.; Guerrero Pantoja, L.; Mena, N.L.; Machuca-Martínez, F.; Urresta, J. Evaluation of TiO<sub>2</sub> and SnO Supported on Graphene Oxide (TiO<sub>2</sub>-GO and SnO-GO) Photocatalysts for Treatment of Hospital Wastewater. *Water* **2020**, *12*, 1438. [[CrossRef](#)]
6. Karbasi, M.; Karimzadeh, F.; Raeissi, K.; Rtimi, S.; Kiwi, J.; Giannakis, S.; Pulgarin, C. Insights into the photocatalytic bacterial inactivation by flower-like Bi<sub>2</sub>WO<sub>6</sub> under solar or visible light, through in situ monitoring and determination of reactive oxygen species (ROS). *Water (Switzerland) MDPI AG*. *Water* **2020**, *12*, 1099. [[CrossRef](#)]
7. Arce-Sarria, A.; Aldana-Villegas, K.M.; Betancourt-Buitrago, L.A.; Colina-Márquez, J.Á.; Machuca-Martínez, F.; Mueses, M.A. Degradation of Hexacyanoferrate (III) from Gold Mining Wastewaters via UV-A/LED Photocatalysis Using Modified TiO<sub>2</sub> P25. *Water* **2020**, *12*, 2531. [[CrossRef](#)]
8. Graça, C.A.L.; Ribeirinho-Soares, S.; Abreu-Silva, J.; Ramos, I.I.; Ribeiro, A.R.; Castro-Silva, S.M.; Segundo, M.A.; Manaia, C.M.; Nunes, O.C.; Silva, A.M.T. A Pilot Study Combining Ultrafiltration with Ozonation for the Treatment of Secondary Urban Wastewater: Organic Micropollutants, Microbial Load and Biological Effects. *Water* **2020**, *12*, 3458. [[CrossRef](#)]
9. Acosta-Herazo, R.; Cañaverl-Velásquez, B.; Pérez-Giraldo, K.; Mueses, M.A.; Pinzón-Cárdenas, M.H.; Machuca-Martínez, F. A MATLAB-Based Application for Modeling and Simulation of Solar Slurry Photocatalytic Reactors for Environmental Applications. *Water* **2020**, *12*, 2196. [[CrossRef](#)]