Hindawi
Journal of Chemistry
Volume 2022, Article ID 9727857, 24 pages
https://doi.org/10.1155/2022/9727857

Review Article

Emerging Pollutants in Moroccan Wastewater: Occurrence, Impact, and Removal Technologies

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Received 11 October 2021; Revised 7 December 2021; Accepted 27 December 2021; Published 25 January 2022

Academic Editor: Umair Yaqub Qazi

The rapid growth of anthropogenic activities in recent decades has resulted in the appearance of numerous new chemical compounds in the environment, known as "emerging pollutants" (EPs) or "contaminants of emerging concern" (CECs). Although partially or not yet regulated or monitored, there is growing research interest in these EPs among the scientific community because of their bioaccumulation, persistence, and adverse effects. Among these, endocrine disruptors, pesticides, and pharmaceuticals can have harmful impacts on human health and the ecosystem. Conventional wastewater treatment technologies are not effective in removing these contaminants, allowing them to be released into the receiving environment. In order to improve the understanding of emerging pollutants, this review discusses the source, occurrence, and impacts of bisphenol A, atrazine, amoxicillin, and paracetamol as model molecules of emerging environmental pollutants, an issue that remains underrepresented in Morocco. Then, treatment methods for EPs are reviewed, including adsorption, advanced oxidation processes, biodegradation, and hybrid treatment. It is proposed that adsorption and photocatalysis can be used as simple, effective, and environmentally friendly technologies for their removal. Thus, we summarize some of the adsorbent and photocatalyst materials applied in recent work to control these pollutants. Towards the end of this paper, the development of inexpensive and locally available (Morocco) materials to remove these compounds from wastewater is considered.

1. Introduction

Studies on wastewater quality have generally focused on priority pollutants, nutrients, microbial contaminants, heavy metals, and dyes. Recently, a wide range of pollutants (emerging pollutants) have attracted researchers' attention and pose a risk to the environment and human health, namely, compounds generated by applying new technological processes [1, 2]. Emerging pollutants (EPs), also known as emerging contaminants (ECs) or contaminants of emerging concern (CECs), are a set of newly identified natural or synthetic chemical compounds that are not monitored but may enter the environment and harm aquatic life and humans [3–5]. The Journal *Emerging Contaminants* has defined these compounds as "*…chemicals that are not currently (or have been only recently) regulated and about which there exist concerns regarding their impact on human or ecological health.*" [6].

Emerging pollutants (EPs), such as endocrine disruptors, pesticides, pharmaceuticals, and their degradation products, are of growing and global concern. They can have negative impacts on human and ecosystem health. However, their presence in the environment is generally in the low concentration range (from $\mu g\ L^{-1}$ to $ng\ L^{-1}$) [3, 7]. Therefore, numerous studies have been conducted worldwide on the pollution of the aquatic environment by EPs and have confirmed their presence in almost all aqueous media, such as surface water, groundwater, seawater, drinking water, and wastewater [1, 5, 7, 8].

To date, studies on the identification of these pollutants are extremely limited in a number of developing countries, particularly in Africa as well as in other parts of the world, and are relatively new in some of them, due to a variety of factors, including the lack of analytical methods for detecting pollutants in wastewater. Indeed, scientists in these countries need to accelerate research on the presence and fate of emerging contaminants as recalcitrant water-borne pollutants, from their emission to their discharge into the environment, in order to address threats to human health and environment [3].

Numerous treatment technologies for emerging pollutants have been adopted to reduce their impacts. Current treatment processes fall into three categories: physical, chemical, and biological. Techniques such as adsorption, advanced oxidation processes (AOPs), and biological treatment have been explored to counteract the adverse effects of these contaminants [9]. In particular, adsorption treatment is an effective way to remove emerging contaminants that tend to spread in the environment. In addition, photocatalytic degradation is considered a promising technique for mineralizing a large proportion of trace micropollutants using sunlight [10]. However, new treatment technologies are needed to provide high-quality water for human and environmental needs [11].

Morocco is one of the developing countries that has started to address this type of issues. This review aims to present an overview of the state of the art in the Moroccan context concerning emerging contaminants such as endocrine disruptors, pesticides, and pharmaceuticals. The focus is on a general description of the source of four typical EPs (bisphenol A, atrazine, amoxicillin, and paracetamol), their occurrence in the aquatic environment, and their environmental impacts. Then, treatment methods for these compounds, including adsorption, advanced chemical oxidation, biodegradation, and hybrid processes, are reviewed.

The ultimate purpose of this review is to present the core of emerging pollutant treatment via adsorption and photocatalytic degradation, as well as their combination. The research directions in this field should focus on the valorization of natural resources and waste in order to develop highly efficient adsorbents and photocatalysts from locally sourced materials. In addition, solar energy systems should be given special attention in Morocco in order to make more efficient use of these renewable energy sources. Towards the end of the paper, the application of coupled adsorption and photocatalysis for the treatment of emerging pollutants in Moroccan wastewater is considered.

2. Emerging Pollutants (EPs)

Emerging contaminants are typically substances with a newly identified source/alternative route to humans [9]. They can enter the environment from various sources, such as industrial effluents, agricultural runoff, and leaking domestic wastewater and municipal wastewater treatment plants [9, 10]. Figure 1 depicts the pathways of EPs into the aquatic environment. According to previous studies, existing conventional wastewater treatment plants are ineffective in removing/degrading many of these contaminants, allowing them to be released into the environment and threaten living organisms and human health [3, 11].

2.1. Endocrine Disruptors (EDs). Endocrine disruptors are defined as chemical substances of natural or synthetic origin that can interfere with the hormonal system (in which hormones work as chemical messengers to control and coordinate body functions) [7, 12] by altering processes such as the synthesis, storage, release, metabolism, and transport of the body's natural hormones [10]. These are compounds that can accumulate in the environment and have harmful effects on the ecosystem and on human health.

EDs are classically grouped into families according to their use, including pesticides, flame retardants, natural and synthetic hormones, plasticizers, personal care products, detergents, and some pharmaceuticals [13, 14]. They are present in almost all aqueous media at various levels, such as surface water, groundwater, seawater, wastewater from wastewater treatment plants, and drinking water [15]. These components are of great concern because of their negative impact on the ecosystem.

2.2. Pesticides. Pesticides encompass all compounds intended to prevent, destroy, repel, or mitigate pests [16]. Their use can be diverse, being applied on agricultural land, private gardens, and other public spaces [17]. They are widely used in different parts of the world, according to the literature review.

Pesticides are generally classified into four broad categories according to their intended targets: herbicides, insecticides, fungicides, and bactericides [16]. Although applied to the soil, these substances can be transported offfield into water bodies by surface runoff and percolation through the soil, thereby affecting water quality by posing a risk to aquatic compartments and human health [18, 19].

2.3. Pharmaceuticals. Pharmaceuticals are organic compounds used in medicines to prevent and treat disease and protect public health [20]. Medicines for human or veterinary use are increasingly part of everyday life. They can be classified according to their therapeutic category, such as analgesics, antibiotics, anti-inflammatories, antidepressants, lipid-lowering drugs, and beta-blockers [21].

Pharmaceuticals have been found in wastewater effluents, surface water, groundwater, and sea water at different concentrations [22]. They reach water bodies from various sources, such as excretion by the human body (which introduces them into the sewage system), drainage water, or industrial effluents [23, 24]. Some substances are highly soluble in water, and conventional wastewater treatment processes are not designed to remove these pollutants,

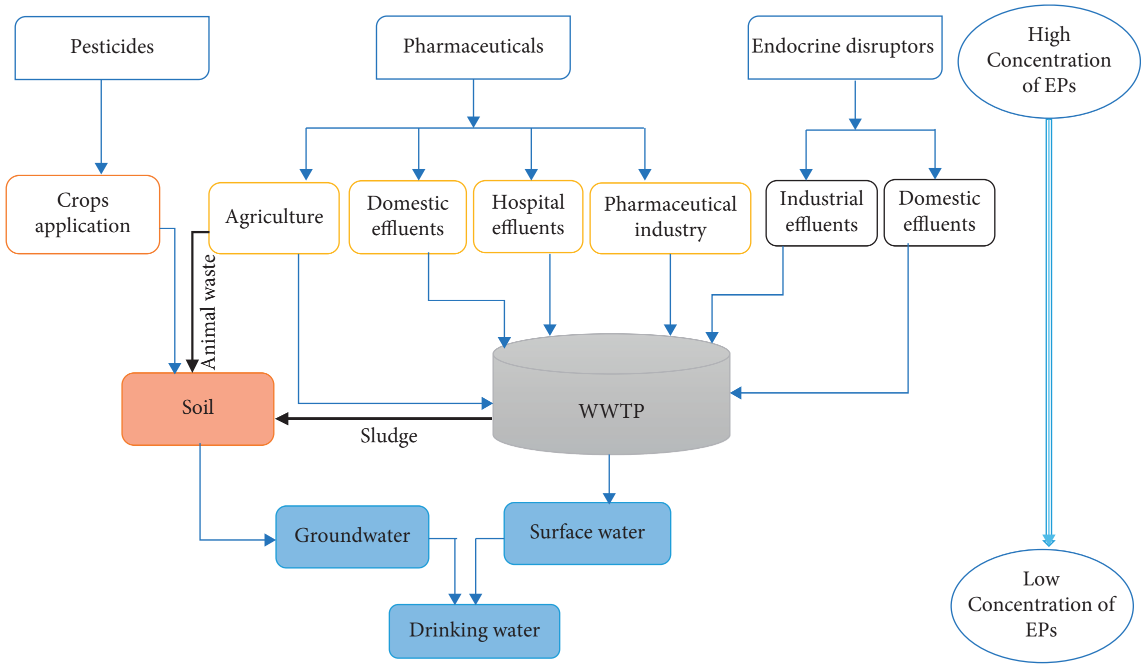


FIGURE 1: Pathways of EPs from the source to the aquatic environment.

allowing them to enter surface water [20]. The widespread presence of these contaminants in the environment has attracted worldwide interest and attention.

3. Representatives of EPs

Among the various emerging pollutants in the environment, bisphenol A (an endocrine disruptor), atrazine (herbicide), amoxicillin (an antibiotic), and paracetamol (an analgesic) were chosen as model molecules for discussion and critical review because of their widespread presence, bioaccumulation, and adverse effects on the aquatic environment and humans. A general description of the applications of these compounds in human life is given below.

3.1. Bisphenol A (BPA). Bisphenol A (BPA, 4, 4′-dihydroxy-2, 2′-diphenylpropane) is a widely used chemical in industry, synthesised by the Russian chemist Aleksandr P. Dianin in 1891 [25]. It is used as an intermediate in the production of polymeric materials such as polycarbonates and epoxy resins. It is present in many products, such as baby bottles, paper, toys, packaging, food containers, paints, medical equipment, and electronics [26, 27]. Bisphenol A is a white crystalline solid with the chemical formula $C_{15}H_{16}O_2$, a molecular weight of 228.29 g mol^{-1}, a low solubility in water (120 mg L^{-1} at 20°C), a log K_{OW} partition coefficient of 3.6, and an acidity constant of 10.3 [28].

BPA is one of the most manufactured and used chemicals in the world. Between 2013 and 2019, global BPA production was found to increase at an annual rate of 4.6% [29]. Studies in several countries have shown the presence of BPA in surface water, groundwater, wastewater, and sludge [29, 30]. It is introduced into the environment from various sources, such as sewage plant effluents, landfill leachate, and industrial discharges [31, 32]. Due to its endocrine-disrupting properties, BPA is considered an environmental contaminant of concern. Its use has been restricted in many countries, such as the European Union, North America, Norway, and China [29, 33].

3.2. Atrazine (ATZ). Atrazine (2-chloro-4-ethylamino-6-isopropylamino-1, 3, 5 triazine) is the active substance of a plant protection product belonging to the triazine chemical family which has a herbicidal effect. It is widely used in agriculture to control weeds in various crops, such as maize, sorghum, and sugarcane [34, 35]. Atrazine is a weak base with the chemical formula $C_8H_{14}ClN_5$, a molecular weight of 215.68 g mol^{-1}, and a low solubility in water (35 mg L^{-1} at 25°C) but a high solubility in organic solvents, with a log K_{OW} partition coefficient of 2.61. It exists as colourless crystals [36].

Although produced to control weeds, atrazine can migrate from soils into the aquatic environment, eventually affecting water quality [37]. Atrazine is frequently detected in soil, surface water, and groundwater, and its routes of entry into the environment mainly include runoff, leaching, and precipitation [36, 37]. Atrazine has endocrine-disrupting properties, and it is a carcinogen that can interfere with ecosystems and cause severe risks to humans, animals, and aquatic life [35, 38]. The European Union banned the compound in 2004, and its use is declining in Canada, while it is still used in other countries, including the United States, China, India, and Brazil [39].

3.3. Amoxicillin (AMX). Amoxicillin is a beta-lactam antibiotic belonging to the penicillin class, used to treat microbial infections by inhibiting the growth of protozoa, bacteria, and fungi [39, 40]. Amoxicillin is one of Morocco's most commonly sold and used drugs [41]. Amoxicillin is a white powder with the chemical formula $C_{16}H_{19}N_3O_5S$, a molecular weight of 365.4 g mol^{-1}, a water solubility of 3430 mg L^{-1} at 20°C, and a log K_{OW} partition coefficient of 0.87. Amoxicillin is extremely unstable and degrades rapidly into a variety of degradation products [42].

Antibiotics, including AMX, are among the many emerging pollutants that have been detected in various environmental matrices, including rivers, groundwater, drinking water, and wastewater treatment plants [41, 43]. They are released into the environment from the pharmaceutical industry, hospital effluents, livestock waste, and sewage effluents [42, 44]. AMX can cause the selection of antibiotic-resistant bacteria and toxicological problems in the aquatic environment, impacting aquatic life and other organisms [45–47]. The widespread use of antibiotics has attracted scientific attention in recent years because of their subsequent release into the environment.

3.4. Paracetamol (PCM). Paracetamol (N-(4-hydroxyphenyl) acetamide), also known as acetaminophen, is a chemical compound used as an analgesic (painkiller) and antipyretic (fever reducer) and is one of the most commonly used and prescribed medicines in the world [48]. In Morocco, it is ranked as the most sold drug [41]. It is recommended as a first-line treatment to reduce fever caused by COVID-19 [49]. Paracetamol is a weak acid with the chemical formula $C_8H_9NO_2$, a molecular weight of 151.16 g mol^{-1}, a high solubility in water (14000 mg L^{-1} at 20°C), and a log K_{OW} partition coefficient of 0.46. It is a white crystallized powder [42]. Paracetamol is capable of being converted into a toxic substance.

Due to its extensive use, its presence has been detected in various environmental matrices, namely, wastewater, surface water, groundwater, sludge, and sediments [38]. The presence of medicines (including paracetamol) in the environment is due to different sources: humans (excretion of drugs or their metabolites that are not absorbed by the human body), industry (manufacturing residues of pharmaceuticals), and agriculture (animal husbandry) [50].

Although the concentration of these substances in aquatic environments is in the range of ng L^{-1} to μg L^{-1}, the continued consumption of these low concentrations can have adverse effects on human health and aquatic organisms [51]. Indeed, the significant presence of traces of these pollutants and, in particular, their metabolites in the environment has become a significant concern, requiring the development of specific innovative treatment techniques to reduce their harmful effects. Table 1 briefly summarizes the physicochemical properties of bisphenol A, atrazine, amoxicillin, and paracetamol.

4. Occurrence in the Environment

Several studies have investigated the presence and effects of emerging pollutants in the aquatic environment, such as the toxicity of bisphenol A and atrazine and the accumulation of certain pharmaceuticals (e.g., amoxicillin and paracetamol), as their concentrations at a certain location and over time differ considerably.

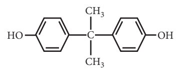
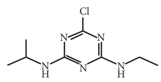
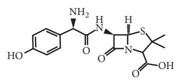
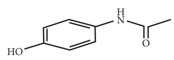
The occurrence of bisphenol A, atrazine, amoxicillin, and paracetamol in surface water, groundwater, wastewater, and seawater from 11 countries was examined in this study, and the results are summarised in Table 2. For instance, Chaib et al. [52] determined the presence of seven antibiotics in the Sebou River (surface water) of the city of Fez (Morocco), in which amoxicillin had the highest concentration (C_{Max} = 4107 ng L^{-1}). In contrast, Radwan et al. [53] found phenolic EDs in surface and groundwater in Egypt and bisphenol A was most frequently detected in surface water with the maximum (C_{Max}) and average (C_{Avg}) concentrations (85,500 and 1085.3 ng L^{-1}, respectively), followed by methylparaben in groundwater with a C_{Max} of 680 ng L^{-1} and a C_{Avg} of 71.1 ng L^{-1}; these results show that bisphenol A is present at high concentrations in water resources in Egypt.

The herbicide atrazine and its metabolites desethylatrazine and hydroxyatrazine were detected in shallow groundwater in the Chesapeake Bay catchment at different concentrations (13.5, 6.5, and 8.5 ng L^{-1}, respectively) [59]. ATZ was also present in all samples and is the predominant pesticide, with detection frequencies of 100%, in the Liaodong Peninsula (China) [54]. As for surface water and groundwater, seawater pollution is the highest in the Eastern Mediterranean Sea (Saronikos Gulf and Elefsis Bay in Central Aegean). Alygizakis et al. [63] investigated the presence of 158 pharmaceuticals and drugs in seawater, in which amoxicillin was the compound detected at the highest levels (up to 127.8 ng L^{-1}). The source of these pollutants is treated wastewater from the greater Athens region, as this area is affected by various anthropogenic pressures.

Indeed, wastewater treatment plants (WWTPs) are considered the main pathway for the transfer of micropollutants to the aquatic environment. The finding of Palli et al. [56] stated nine pharmaceuticals belonging to different therapeutic groups in a campaign conducted in 2017; in the influents of WWTPs in Tuscany (Italy), paracetamol and amoxicillin were frequently detected at higher concentrations (3914 and 2002 ng L^{-1}, respectively). Furthermore, Xue and Kannan [57] studied the presence of eight bisphenols in the final effluent of the Albany Area WWTP (USA). BPA was the most detected compound, at an average concentration of 90.0 ng L^{-1}, and incomplete removal of these compounds was observed, with the highest removal rate (52%) being after secondary treatment. Currently, conventional wastewater treatment processes have proven to be less effective in removing emerging micropollutants. Therefore, they are the main supplier of these pollutants to the environment.

The monitoring and determination of emerging pollutants in aqueous samples involves different analytical techniques. In general, an analytical procedure consists of four main steps: sample preparation, extraction, separation, detection, and quantification (Figure 2) [7]. Most samples require preparation so that they can be easily processed. Liquid samples are often concentrated first with solid adsorbents and then eluted with a suitable organic solvent or

Table 1: Physicochemical properties of bisphenol A, atrazine, amoxicillin, and paracetamol.

Compounds	Chemical formula	Chemical structure	CAS number	Molecular weight (g/mol)	Water solubility (mg/L)	Log K_{ow}	Pk_a	References
Bisphenol A	$C_{15}H_{16}O_2$		80-05-7	228.29	120 (at 20°C)	3.60	10.30	[28]
Atrazine	$C_8H_{14}ClN_5$		1912-24-9	215.68	35 (at 25°C)	2.61	—	[36]
Amoxicillin	$C_{16}H_{19}N_3O_5S$		26787-78-0	365.4	3430 (at 20°C)	0.87	3.20	[42]
Paracetamol	$C_8H_9NO_2$		103-90-2	151.16	14000 (at 20°C)	0.46	9.38	[42]

CAS = chemical abstracts service; log K_{ow} = octanol-water partition coefficient; pK_a = acid dissociation constant.

Table 2: Occurrence of emerging pollutants in the aquatic environment.

Type of water	Sample (country)	Pollutants	Concentration (ng L^{-1})	Extraction and detection methods	LODs/LOQs* (ng L^{-1})	References
Surface water	Sebou River, Morocco	Amoxicillin	<158.3–4107	SPE-LC-MS/MS	n. a	[52]
	Nile River, Egypt	Bisphenol A	1085.3	UPLC-MS/MS	n. a	[53]
	Liaodong Peninsula, China	Atrazine	8.7–64.8	SPE-TQMS	(0.1–1.5)*	[54]
	Al-Asfar and Al-Hubail lakes, Saudi Arabia	Paracetamol and bisphenol A	PCM = 105–3069 BPA = 484.9	SPE-UHPLC/MS/MS	(0.3–2.5)*	[55]
Wastewater	WWTP of Tuscany, Italy	Paracetamol and amoxicillin	PCM = 3914 AMX = 2002	SPE-LC/MS	n. a	[56]
	Albany area of New York State (final effluent), USA	Bisphenol A	49.9	SPE-HPLC/MS/MS	n. a	[57]
	Grahamstown wastewater, South Africa	Bisphenol A	1468.3	SPE-UHPLC/MS/MS	1.0	[58]
Ground water	Groundwater of Chesapeake, USA	Atrazine, deethylatrazine (DEA), and hydroxyatrazine (HA)	ATZ = 13.5 DEA = 6.5 HA = 8.3	LC/MS/MS	n. a	[59]
	Shallow Nile aquifers, Egypt	Bisphenol A	71.1	UPLC-MS/MS	n. a	[53]
	Groundwater of Lagos, Nigeria	Amoxicillin and paracetamol	44–6490 1–188	UPLC-HRMS	n. a	[60]
Sea water	Marine waters, Turkey	Bisphenol A	4160–16920	SPE-HPLC/FLD	n. a	[61]
	Marine waters, Portugal	Paracetamol	53.2–269.7	SPE-UHPLC/MS/MS	0.26/0.80*	[62]
	Eastern Mediterranean Sea, Greece	Amoxicillin	<5.0–127.8	UHPLC/MS	8.2	[63]

LODs: limit of detections, LOQs: limit of quantifications, SPE: solid phase extraction, LC: liquid chromatography, MS: mass spectrometry, HPLC: high-performance liquid chromatography, UPLC: ultraperformance liquid chromatography, UHPLC: ultra-high-performance liquid chromatography, FLD: fluorescence detector, TQMS: triple quadrupole mass spectrometer, HRMS: high-resolution mass spectrometry, n. a: not available.

solvent mixture [64]. For extraction, there are many techniques such as solid phase extraction (SPE), liquid-liquid extraction (LLE), and accelerated solvent extraction (ASE). The choice of a specific extraction technique depends largely on the types of samples, the analyte, and the desired turnaround time [64]. In addition, separation, detection, and quantification are usually performed by liquid or gas chromatography in most cases, coupled with mass spectrometry (MS), due to the high selectivity and sensitivity they offer [65]. Further details on analytical techniques for emerging pollutants in water can be found in the following publications [66, 67].

5. Environmental Impact of EPs

5.1. On the Worldwide Level. The continuous entry of emerging pollutants into the environment is increasingly affecting the quality of water resources, fauna, and flora.

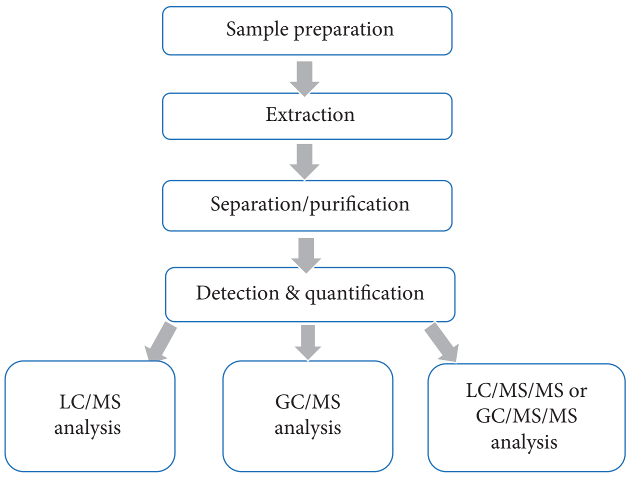


Figure 2: Typical analytical procedure for the measurement of emerging pollutants in aqueous samples.

Several studies conducted worldwide have focused on these pollutants to better understand their toxicity, environmental impact, and behaviour in different aquatic environments. Contaminants from industrial additives, pesticides, and pharmaceuticals, which are persistent compounds, enter water bodies from a variety of sources and can exceed acceptable levels and accumulate, resulting in adverse effects on the environment and human communities [3, 8, 11].

As illustrated in Figure 3, emerging pollutants have a significant effect on humans and animals. One of the consequences of endocrine disruption is that it affects on the endocrine system by mimicking, suppressing, or altering the function of hormones [68]. Exposure to EDs has been shown to reduce male sperm count and increase testicular, prostate, ovarian, and breast cancer, as well as reproductive malfunctions [15, 68]. Moreover, some of the most noticeable effects of EDs on animals are the changes in reproductive anatomy, fertility, eggshell thinning, and hormonal activity [70]. Pharmaceuticals and pesticides can also have an effect on the growth, reproduction, and evolution of species in the environment.

For instance, bisphenol A, a chemical widely used in the manufacture of plastics, has been confirmed to act as an endocrine disruptor, affecting organs such as the thyroid, thymus, and pancreas, due to its ability to bind to various receptors associated with the endocrine system, thereby disrupting their functions [56, 63]. BPA has adverse effects on plants, animals, and humans. Some studies have indicated that exposure to BPA causes numerous endocrine, reproductive, and metabolic diseases in humans, including breast, endometrial, prostate, ovarian, and testicular cancers, diabetes, obesity, hypertension, and heart disease [57, 64]. Several countries have banned its use in consumer products in response to these concerns, including baby bottles [30].

The effects of BPA on animals revealed a decrease in the number of elongated spermatids in the seminiferous tubules of pubertal ICR mice and a decrease in sperm count in Holtzman rats [71]. Additionally, the effects on fish have been investigated, including transcriptional activation of estrogen receptor-responsive genes, increased brain aromatase activity, induction of VTG in males, disruption of gametogenesis in males and females, altered development (neuronal, cardiac, germ cell, and sexual differentiation), and changes in sex ratios following embryonic exposure [72].

On the other hand, atrazine is a herbicide applied in various crops to control weeds. However, this compound can also directly or indirectly affect other organisms [37]. In aquatic ecosystems, atrazine can have harmful effects on aquatic animals and plants. Many organs, such as the kidneys, liver, gills, and other organs of fish, are affected after exposure [35]. Atrazine is an endocrine-disrupting compound that can also alter male reproductive tissue when animals are exposed to it during development [73]. Moreover, it was discovered that ATZ induced DNA methylation in the carp brain and induced autophagy in the liver, and it can also affect human health through the skin and respiratory contact, causing ovarian and breast cancer and affecting the human vascular system [74].

Pharmaceuticals are a specific class of compounds used worldwide to treat disease and restore the health of the body [75]. Still, they can also have harmful effects and contribute to pollution. In the aquatic environment, amoxicillin (an antibiotic) creates an ecological imbalance by producing toxic effects on aquatic organisms, altering plant growth, causing abnormalities in the anatomy of many organisms [41, 43], and leading to the development of multidrug-resistant bacteria [76]. Amoxicillin is a potential mutagen, carcinogen, and teratogen at higher doses; it is toxic to the fish *Oryzias latipes* and has a 96 h LC50 of 1000 mg L^{-1} [77].

Paracetamol has adverse effects when overdosed, inducing the proliferation of breast cancer cells. This toxicity is typically attributed to reactive oxygen species, which cause a variety of effects ranging from protein denaturation to lipid peroxidation and DNA damage [78]. PCM was also evaluated for its toxicity against a variety of aquatic species, including bivalve species (*C. fluminea*) and crustaceans (*Daphnia magna*). Exposure to increasing concentrations significantly altered the redox status of *C. fluminea*. Likewise, it caused death in a chronic toxicity experiment with *Daphnia magna* at higher concentrations (1.2–1.7 mg L^{-1}) [79].

5.2. At the National Level (Morocco). Due to population growth and the increase in agricultural and industrial activities, Morocco's water resources are subject to increasing and continuous pressure that affects their quality. The aquatic environment remains the most affected due to wastewater discharge containing industrial products, pharmaceuticals, and others, which are incompletely eliminated by treatment plants and seriously pollute the water bodies.

The 3rd report entitled "The state of the environment in Morocco, 2015" [80], from the Ministry of Energy, Mines, Water, and the Environment, in charge of the environment, provides information on the main activities causing pollution of the aquatic environment: the industrial sector accounts for a considerable proportion of polluting emissions.

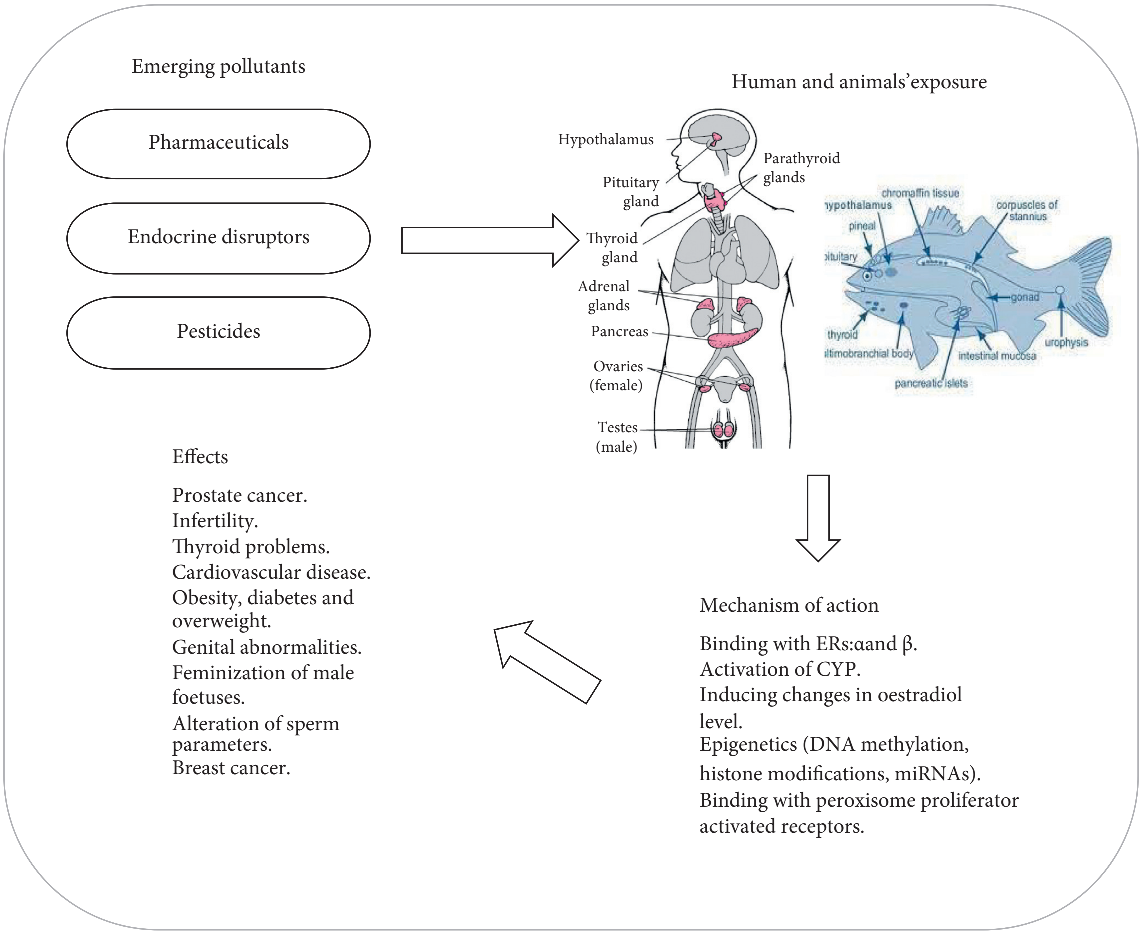


FIGURE 3: Schematic representation of emerging pollutant exposures, mode of action, and their health effects (based on [25, 68, 69]). Abbreviations: ERs, estrogen receptors; CYP, cytochrome P450 genes; and DNA, deoxyribonucleic acid.

The production of industrial wastewater in Morocco is around 964 million m^3 per year, i.e., 92% of the water initially taken from the sea and freshwater resources. The chemical and parachemical industries are the main sources of wastewater discharge, with an annual volume of 931 million m^3, while the other sources with an impact on the volume of wastewater are the agroindustry, the textile and leather sectors, and, to a lesser extent, the mechanical and metallurgical industries.

Furthermore, urban wastewater discharge is also a major source of pollution, with the annual volume of wastewater discharged into the natural environment (sea, rivers, sewers, etc.) without prior treatment estimated at 870 million m^3 in 2020 and predicted to be 1039 million m^3 in 2030. For example, the two main rivers of Morocco, the Sebou and the Oum Er Rbia, regularly experience critical pollution situations lasting several years. Additionally, the agricultural sector consumes an excessive amount of water (85–90% of resources absorbed by agriculture). Therefore, the leaching of phytosanitary products used in agriculture may also lead to water contamination by pesticides [81].

These different and large quantities of effluents released into the environment may contain varying concentrations of emerging pollutants, including pesticides, industrial products, and pharmaceutical products. For instance, an integrated study by Azzouz et al. [82] showed the presence of six types of endocrine disruptor (including BPA) in fish and seafood samples from Europe and North Africa. This study shows the consumption of these pollutants by aquatic organisms.

The finding of Chafi et al. [83] indicated that 27 endocrine disruptors and pharmaceuticals are present in Moroccan surface waters (Bouregreg River), in which bisphenol A and paracetamol are presented with maximum concentrations (302 and 120 ng L^{-1}). On the other hand, Chaib et al. [52] studied the occurrence of amoxicillin and other antibiotics in the surface water of the city of Fez (Morocco), where amoxicillin concentration reached 4107 ng L^{-1}. Indeed, few research studies have been conducted in Morocco on the effects of these contaminants on human health and the environment. Therefore, further research on this aspect is needed.

6. Treatment Processes for EP Removal

Varying levels of emerging pollutants in water matrices have attracted the interest of scientists around the world. There is, therefore, a need to protect humans and the environment from these contaminants and their impact. Emerging pollutants can be removed from water by chemical, physical,

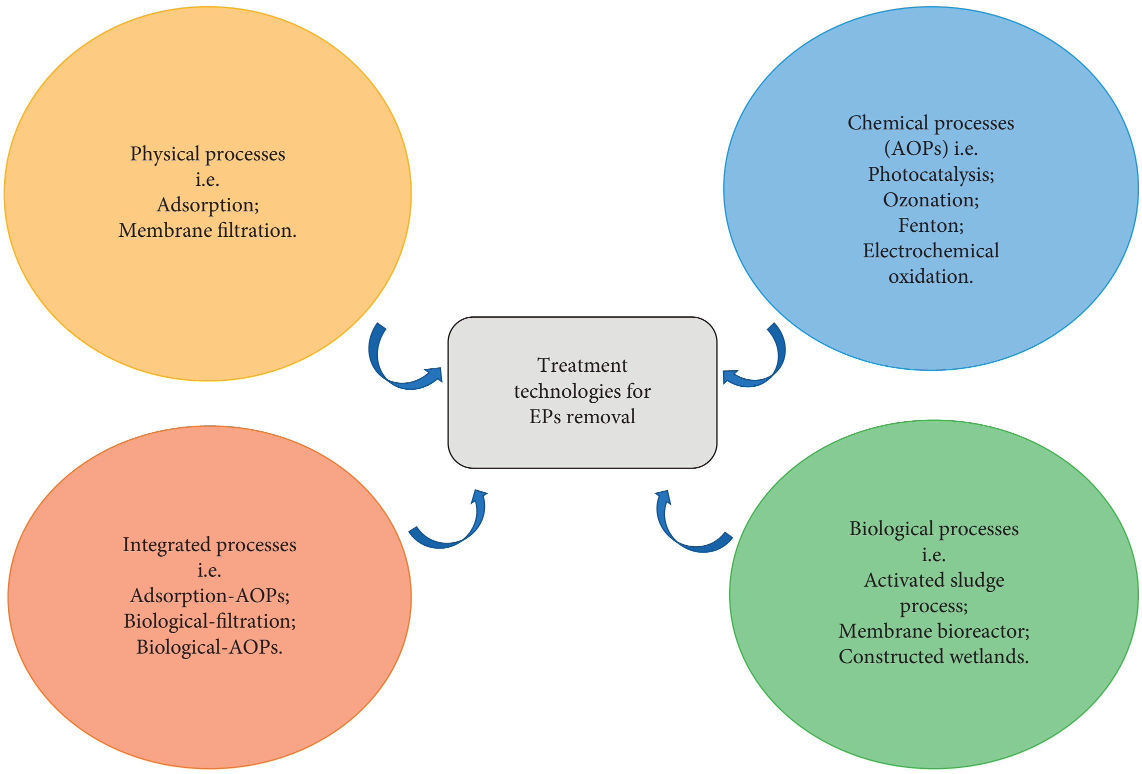


Figure 4: Common treatment technologies for EP removal.

biological, and hybrid processes. The most common technologies for their removal are shown in Figure 4.

Chemical treatment generally means the use of chemicals in a series of reactions to facilitate the process of disinfecting wastewater, and the chemical oxidation method has been employed to convert pollutants into a harmless form [68]. Thus, physical treatment techniques are frequently used due to their simplicity and adaptability. Commonly used physical processes include adsorption and membrane technologies, such as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis. In addition, biological treatment uses a variety of micro-organisms to remove contaminants by biodegradation. The main nonconventional biological processes are activated sludge processes, constructed wetlands, and membrane bioreactors (MBRs).

Hybrid wastewater treatment is a system that integrates two or more treatment processes, in which all chemical, physical, and/or biological treatment techniques are integrated to facilitate the removal of contaminants from wastewater. The following sections of the document provide a more detailed overview of treatment techniques that have been applied to reduce the effects of these contaminants in wastewater. These methods are adsorption, advanced oxidation processes, biological processes, and hybrid (integrated) processes.

6.1. Adsorption. Adsorption is a mass transfer process that involves the mass movement of the adsorbate from the liquid or gas phase to the surface of the adsorbent [84]. Adsorption mechanisms generally include physical adsorption related to van der Waals force and ion exchange and chemical adsorption corresponding to the formation of chemical bonds [85]. Adsorption is a widely applied technology for the removal of various contaminants from water and wastewater because it is simple, effective, cheap, and environmentally friendly [80, 81]. This process can be influenced by many conditions, such as the nature and concentration of the adsorbate and the adsorbent, the presence of other pollutants, and temperature and experimental parameters such as contact time, pH, and adsorbent surface [15].

Many researchers have explored the removal of emerging pollutants by adsorption. Activated carbon is the best and most widely used adsorbent globally due to its high efficiency in removing different types of contaminants. However, it is expensive and difficult to regenerate, which requires the search for other materials with similar efficiency [81]. Other materials have been studied as alternatives to activated carbon, such as activated biochar, clay, zeolites, silica gel, chitosan, metal-organic frameworks, polymers, agricultural waste and by-products, biosorbents, and composite materials. Table 3 shows some adsorbent materials that have recently been applied to remove the studied pollutants from aqueous solutions.

Many studies have been conducted on the adsorption of emerging pollutants by different materials. Some studies have used carbon-based adsorbents to remove several types of pollutants. Zbair et al. [86] studied the adsorption of bisphenol A in water by activated carbon prepared from argan nut shells and activated by phosphoric acid, removing 1250 mg/g BPA. The optimized variables were 0.01 g, 60 mg L^{-1} and 6.5 as adsorbent dosage, initial BPA concentration,

Table 3: Removal efficiency, isotherms, and kinetic parameters for the removal of the studied emerging pollutants by different adsorbents.

Adsorbate	Adsorbent	Surface area (m^2/g)	Removal (%)	Removal Q_m (mg/g)	Adsorption isotherm	Operating conditions	Additional information	References
Bisphenol A	Activated carbon from argan waste	1372	95	1250	Langmuir	200 mL of BPA (60 mg/L); $m = 0.01$ g of adsorbent; PH = 6.5; $T = 293$ K; $t = 3$ h; SS = 200 rpm; pseudo-second-order	Activated carbon prepared from the shell of the argan nut and activated by phosphoric acid	[86]
	Activated biochar from kraft lignin	1053		220	Dual-site Langmuir	[BPA] = 100 mg/L; [adsorbent] = 0.36 g/L; $T = 25°C$; $t = 24$ h; SS = 250 rpm; Elovich model	Activated carbon from kraft lignin showed a high BPA uptake value in a batch experiment with synthetic wastewater	[87]
	Clay	15.74		109.89	Langmuir and Freundlich	[BPA] = 50 mg/L; PH = 7; $T = 25°C$ $t = 4$ h; pseudo-second-order	Batch experiment was carried out to determine the adsorption characteristics of calcium-modified montmorillonite clay towards BPA	[88]
	Graphene oxide (GO)	—	96.2	3293.9	Sips	30 mL of BPA (1 mg/L); $m = 2.5$ mg of adsorbent; PH = 7; $T = 25°C$; $t = 120$ min; pseudo-second-order; $K_2 = 25.2$ g mg^{-1} min^{-1}	The hybrid of GO with Fe_2O_3 nanoparticles (Fe_2O_3-GO) had a higher adsorption at a lower initial BPA concentration, batch experiment with synthetic wastewater	[89]
	Polymer	—		65.3	Langmuir	20 mL of BPA (100 mg/L); $m = 5$ mg of adsorbent; PH = 7; $T = 25°C$; SS = 200 rpm; pseudo-second-order	Synthesis of a water-insoluble polymer (b-PEI-PEG-β-CD) that could easily remove BPA from synthetic wastewater	[90]
	Sulfonated tea leaves	—		236.8	Langmuir	20 mL of BPA (100 ppm); $m = 10$ mg of adsorbent; $T = 25°C$; PH = 8; SS = 700 rpm; pseudo-second-order; $K_2 = 0.000356$ g/mg min	Sulfonation of tea leaves generates the sulfonated carbonaceous product TW-SO_3H with high adsorption capacity towards BPA	[91]

Table 3: Continued.

Adsorbate	Adsorbent	Surface area (m^2/g)	Removal (%)	Removal Q_m (mg/g)	Adsorption isotherm	Operating conditions	Additional information	References
Atrazine	Metal-organic frameworks (MOFs)	2210	98	36	Langmuir	10 mL of ATZ (10 ppm); $m = 3.5$ mg of adsorbent; $T = 25°C$; $t = 1$ min	Adsorption of atrazine in Zr_6-based metal-organic structures showed a high adsorption capacity (98%) in 1 minute	[92]
	Polyaniline-derived carbon	—		943.0	Langmuir	100 mL of ATZ (50 mg/L); $m = 3$ mg of adsorbent; $T = 25°C$; PH = 7; $t = 12$ h	Preparation and use of polyaniline carbons for the adsorptive removal of ATZ from synthetic wastewater	[93]
	Biosorbent from eucalyptus bark	—	87.95	936.1	Freundlich	10 mL of ATZ (1 mg/L); $m = 30$ mg of adsorbent; $T = 25°C$; $t = 24$ h; SS = 225; pseudo-second-order	*Eucalyptus tereticornis* L. bark, a waste product, is used to remove atrazine in a batch adsorption experiment	[94]
	Biochar	—	96	79.6	Freundlich	10 mL of ATZ (2 mg/L); $m = 50$ mg of adsorbent; $T = 25°C$; $t = 20$ min; pseudo-second-order	P-doped biochar from corn straw, prepared and activated with H_3PO_4, was able to remove 96% of atrazine	[95]
Amoxicillin	Activated carbon from date pits	1325		424.3	Langmuir	10 mL of AMX (100 mg/L); $m = 10$ mg of adsorbent; $T = 22°C$; PH = 4; $t = 300$ min; pseudo-second-order	Activated carbon is derived from date pits and prepared by thermal activation with carbon dioxide, used for the removal of amoxicillin in a batch adsorption experiment	[96]
	Natural phosphate	20		3.2	—	100 mL of AMX (20 mg/L); $m = 200$ mg of adsorbent; $T = 25°C$; PH = [5–6]; $t = 120$ min	Natural phosphate from the sedimentary phosphate rocks of Morocco	[97]
	Multiwall carbon nanotubes	—		159.4	Langmuir	100 mL of AMX (50 mg/L); $m = 0.1$ g of adsorbent; $T = 60°C$; PH = 7; $t = 75$ min; pseudo-second-order	Multiwalled carbon nanotubes are used as an adsorbent for the removal of amoxicillin from an aqueous solution in a batch experiment	[98]
	Activated carbon	807	76		Langmuir	2000 mL of AMX (40 mg/L); $m = 2$ g of adsorbent; $T = 25°C$; PH = 6.9; $t = 30$ min; SS = 300 rpm; pseudo-second-order	Activated carbon modified with zinc acetate and activated with phosphoric acid was used in a batch adsorption experiment	[99]
	Modified clay	242.36		647.7	Langmuir	20 mL of AMX (50 mg/L); $m = 2$ mg of adsorbent; $T = 30°C$; PH = 7.5; $t = 60$ min; SS = 120 rpm; pseudo-second-order	Montmorillonite clay modified with L-methionine amino acid was used for amoxicillin adsorption	[100]

Table 3: Continued.

Adsorbate	Adsorbent	Surface area (m^2/g)	Removal (%)	Removal Q_m (mg/g)	Adsorption isotherm	Operating conditions	Additional information	References
Paracetamol	Commercial activated carbon	983		560	Langmuir	[PCM] = 50 mg/L; [adsorbent] = 167 mg/L; T = 25°C; PH = 3; t = 24; SS = 250 rpm; pseudo-second-order	Commercial activated carbon was used for the adsorptive removal of paracetamol in a batch adsorption experiment	[48]
	Modified clay	216		22.08	Redlich–Peterson	50 mL of PCM (100 mg/L); m = 5 mg of adsorbent; T = 25°C; PH = 7; t = 180 min; pseudo-second order	Natural montmorillonite clay pillared with titanium oxide	[101]
	Coffee-based biomaterial	888.1	98	50	Freundlich	[PCM] = 200 mg/L; [adsorbent] = 4 g/L; PH = 6.5; t = 60 min; pseudo-second-order	The raw biomaterial treated chemically by phosphoric acid	[102]
	Silica gel	264		95	Langmuir	[PCM] = 100 mg/L; [adsorbent] = 167 mg/L; T = 25°C; PH = 3; t = 24 h; SS = 250 rpm; pseudo-second-order	Removal of paracetamol by silica gel in a batch adsorption experiment	[48]

and pH. Adsorption experiments revealed that the adsorbent was more efficient due its large specific surface area (1372 m^2/g). The obtained adsorption data were highly correlated with the pseudo-second-order model and the Langmuir isotherm.

Meanwhile, Suo et al. [95] found that P-doped biochar from maize straw was able to remove 96% of atrazine within 20 min and at a temperature of 25°C. The experimental data were best fitted by a pseudo-second-order kinetic model and the Freundlich isotherm. Additionally, the prepared adsorbent can be reused up to five cycles. There are other studies on the adsorption of these pollutants by mineral adsorbents; Chauhan et al. [101] investigated the removal of paracetamol by natural montmorillonite clay coated with titanium oxide and found a lower adsorption capacity of 22.08 mg/g at a fixed adsorbent dose (0.1 g L^{-1}). Pseudo-first-order and pseudo-second-order models were used to determine the adsorption kinetics.

Rock phosphate from sedimentary phosphate rocks in Morocco was used to remove amoxicillin and other organic micropollutants. The optimized parameters for contact time, adsorbent concentration, pH, and initial AMX concentration were 120.0 min, 2.0 g L^{-1}, 6.0, and 20 ppm, respectively. The mechanism of adsorption was related to the nature of van der Waals interactions [97]. Atrazine adsorption in Zr6-based metal-organic frameworks (MOFs) showed a high adsorption capacity (98%) within 1 minute, owing to the framework's large pores that facilitate diffusion and the abundance of potential π–π interaction sites at the pyrene-based linkers. Additionally, the adsorbent was easily regenerated following atrazine adsorption using acetone washing while retaining 99% of its initial atrazine uptake [92].

Other researchers have used waste materials as low-cost adsorbents. Mandal and Singh [94] studied the removal of atrazine by eucalyptus bark in a batch adsorption experiment. The removal efficiency showed that 30 g L^{-1} of adsorbent was capable of removing up to 87.95% of atrazine at a concentration of 1 mg L^{-1}. Kinetic analysis of the equilibrium data indicated that atrazine sorption was best explained by a pseudo-second-order kinetic model. Moreover, sulfonation of tea leaves generates the sulfonated carbon product TW-SO_3H, which has a high adsorption capacity for BPA (236.80 mg g^{-1} at 25°C). The adsorbent exhibited electrostatic interaction and π–π stacking properties that enabled efficient BPA adsorption. The Langmuir and Temkin isotherm models best fit the experimental data for the BPA adsorption processes [91].

Numerous factors can affect the adsorption process, including adsorbent dosage, pollutant concentration, solution pH, contact time, temperature, the nature of the adsorbent and adsorbate, and the presence of other pollutants [84]. The transfer of pollutants in aqueous media can be understood by considering the adsorption isotherm, kinetic, and thermodynamic studies. Adsorption isotherms can be used to determine the mass of adsorbate that is taken onto the surface or interface of an adsorbent at a given temperature and equilibrium. Numerous adsorption isotherm models (Langmuir, Freundlich, Koble–Corrigan, and others) have been proposed to account for the adsorption capacity of pollutants on the adsorbent [81, 84].

Furthermore, adsorption kinetic study provides detail on the rate and mechanism that governs the adsorption phenomenon. The main models used in the literature are the pseudo-first-order kinetic, the pseudo-second-order kinetic, the Elovich model, and the intraparticle diffusion kinetic model [103]. Besides, thermodynamic study elucidates the nature of the adsorption process whether it is of physical/chemical or endothermic/exothermic or spontaneous. The obtained values of entropy change (ΔS^0), Gibbs function change (ΔG^0), enthalpy change (ΔH^0), and activation energy (Ea) can be used to infer the spontaneity of the adsorption process and the adsorption interaction's exothermic/endothermic behavior [84]. Due to its multiple advantages, the adsorption technique is one of the most effective and widely applicable low-cost methods for the treatment of emerging pollutants. Therefore, additional effective and low-cost materials (adsorbents) for wastewater treatment are required.

6.2. Advanced Oxidation Processes (AOPs). AOPs are a family of technologies based on the production of hydroxyl radicals (OH^*), which are stronger oxidants (oxidation potential of 2.8 V) and capable of reacting rapidly with most organic compounds present in water and wastewater [104, 105]. The generation of these reactive radicals can be achieved by several processes, including homogeneous and/or heterogeneous phase photocatalytic processes (H_2O_2/UV, O_3/UV, Fe^{2+}/H_2O_2/UV, and TiO_3/UV), homogeneous phase chemical oxidation processes (H_2O_2/Fe^{2+} and H_2O_2/O_3), electrochemical oxidation processes, and sonochemical oxidation processes [106]. AOPs, including Fenton reactions, photocatalytic oxidation, ozonation, and electrochemical oxidation reactions, are effective in removing emerging pollutants that are difficult to treat by means of conventional physicochemical and biological techniques. The efficiencies of some common advanced oxidation processes are presented in Table 4.

In Fenton and Fenton-type reactions, hydroxyl radicals are usually generated by the decomposition of hydrogen peroxide under the action of an iron-rich catalyst ($BaFe_{12}O_{19}$, α-FeOOH, Fe_3O_4, etc.) [104, 105, 107]. The hydrogen peroxide used in the catalytic reaction usually comes from external addition and in situ generation [121]. However, when mixed with an iron (II) (Fe^{2+}) catalyst to form the Fenton reagent, the oxidation potential of (H_2O_2) increases. A mixture of (H_2O_2) and (Fe^{2+}) salts is added directly to the wastewater [122], according to the following reaction:

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + OH^- + OH^*. \qquad (1)$$

Fenton-based processes have been successfully applied to the treatment of various types of Moroccan wastewater, including textile wastewater [123], landfill leachates [124, 125], and emerging pollutants in aqueous solutions. For instance, the electro-Fenton process has been applied to remove moxifloxacin in acidic media at pH 3.0. The mineralization of moxifloxacin was achieved by multiple OH^* attacks with several intermediates formed during the treatment process [126]. Likewise, Rachidi et al. [127] investigated the same process for the removal of the antidepressant sertraline hydrochloride. The maximum degradation occurred at 400 mA with an optimal Fe^{2+} concentration of 0.1 mM.

Photocatalysis is a sustainable technology for treating organic pollutants in wastewater that involves the use of photocatalysts, having the ability of being activated under light irradiation [128]. The photocatalysis technique is based on the reaction between organic pollutants and powerful oxidizing and reducing agents (h^+ and e^-) generated by a light source on the surface of photocatalysts [129]. The typical photocatalytic mechanism involved in the removal of aqueous phase contaminants is depicted in Figure 5.

Titanium dioxide (TiO_2) is the most widely used photocatalyst due to its numerous advantages in the degradation of contaminants. TiO_2 is less efficient in absorbing solar light. Therefore, most of studies focus on its modification through doping with metals (such as Ag^+, Fe^{3+}, and Co^{3+}) and nonmetals (including N, S, F, C, B, and P) to enhance its visible light-absorbing capacity [130]. Moreover, researchers have been attracted to investigate other photocatalytic materials for wastewater treatment applications, including oxides and perovskites (e.g., ZnO, WO_3, V_2O_5, $BiVO_4$, Ag_3VO_4, and $SrTiO_3$), bismuth oxyhalides (e.g., BiOCl, BiOBr, and BiOI), and sulfides (e.g., CdS, ZnS, and MoS_2), as well as various composite materials [131].

Numerous photocatalytic materials have been employed for the treatment of hazardous contaminants in wastewater [132, 133]. Tabasum et al. [134] studied the photocatalytic potentials of graphene oxide-doped metal ferrites (GO-Fe_3O_4 and GO-$CoFe_2O_4$) for acetamiprid degradation. During the first hour of exposure to UV radiation, degradation efficiencies of 90% and 97%, respectively, were achieved. Additionally, the performance of graphene-oxide-based metal ferrites for pesticide pollutant removal was investigated [135]. The composites were found to be highly biodegradable (90%) within 60 minutes of UV degradation. Qureshi et al. [136] synthesized graphene oxide decorated $ZnWO_4$ (GO-$ZnWO_4$) nanocomposites by a hydrothermal process and used for the degradation of a pharmaceutical product (cetirizine hydrochloride) under UV irradiation. The photocatalyst was able to degrade up to 89% of the contaminant in water.

Another semiconducting photocatalyst has been developed and is being applied to the treatment of emerging pollutants, such as zinc oxide-hydroxyapatite (HAp). El Bekkali et al. [137] explored the use of ZnO-HAp for antibiotic removal from contaminated water under UV irradiations. The photodegradation efficiency of the nanocomposites was significantly higher than that of the photocatalytic particles alone. In addition, activated carbon-based coloured titania nanoparticles showed an excellent performance in removing emerging drugs from wastewater, such as amoxicillin and paracetamol, when exposed to visible light [138]. Furthermore, Bougdour et al. [139] used the $S_2O_8^{2-}/Fe^{2+}$/UV process to investigate the treatment and mineralization of real wastewater from the Moroccan textile industry. The results indicated that the rate of pollutant mineralization is 87%.

Ozonation processes are based on the use of ozone, which is a powerful oxidizing agent. After reacting with the pollutants, ozone is transformed into oxygen [140]. This process has shown relative efficiency in treating emerging pollutants; 100% removal was achieved for bisphenol A in real secondary wastewater effluents [113]. Similarly, total degradation and 94% mineralization of paracetamol were achieved for reaction times of 15 minutes [116]. Likewise, microbubble ozonation improved the degradation of atrazine (90%) at different pH levels in a semibatch experiment [114]. Electrochemical oxidation is mainly based on electron

Table 4: Advanced oxidation processes for the degradation of emerging pollutants present in water.

Process	Target compound	Materials	Degradation (%)	Conditions	Additional information	References
Fenton/Fenton-like processes	Bisphenol A	$BaFe_{12}O_{19}$-Ag_3PO_4	79.9	[BPA] = 20 mg/L; [catalyst] = 1 g/L 300 W; Xe arc lamp ($\lambda > 420$ nm); $T = 30°C$; $t = 30$ min	The reactive oxygen species are produced by the timely decomposition of H_2O_2 generated on the surface of Ag_3PO_4 via the $BaFe_{12}O_{19}$	[107]
	Atrazine	α-FeOOH	100	[ATZ] = 20 mg L^{-1}; [α-FeOOH] = 0.1 g L^{-1}; [H_2O_2] = 1.0 mM; [hydroxylamine] = 0.5 mM; $T = 25°C$; PH = 5; $t = 60$ min	Surface Fenton system constructed with hydroxylamine (NH_2OH), goethite (α-FeOOH), and H_2O_2 (α-FeOOH-HA/H_2O_2) to degrade atrazine	[104]
	Amoxicillin	TiO_2-GO-Fe_3O_4	90	[AMX] = 10 mg/L; [catalyst] = 0.5 g L^{-1}; $t = 120$ min; PH = 3; $T = 25°C$	The combination of TiO_2 and Fe_3O_4 nanoparticles on graphene oxide (GO) nanoplatelets (TiO_2-GO-18wt% Fe_3O_4) shows excellent AMX degradation under visible irradiation and 90% TOC removal	[105]
	Paracetamol	Fe_3O_4-SiO_2-Cu	100	[PCM] = 2.0 mg L^{-1}; [catalyst] = 0.2 g L^{-1}; [H_2O_2] = 15 mM; $t = 20$ min PH = 5.0; $T = 25°C$	The catalytic tests were carried out in a four-vial collared reactor equipped with a temperature-controlled heating mantle; paracetamol was almost completely degraded within 20 minutes	[108]
Photocatalysis	Bisphenol A	B-TiO_2-graphene oxide	47.66	[Catalyst] = 1000 mg L^{-1}; [BPA] = 10 mg L^{-1}; $t = 240$ min; PH = 5; $T = 25°C$; $K_1 = 0.0023$ min^{-1}	Hydrothermal preparation of the photocatalyst, 300 W xenon lamp (1000 W/m^2 light intensity) with solar irradiation	[109]
	Atrazine	Bi_2MoO_6/PMS	99	[Bi_2MoO_6] = 0.6 g/L; [PMS] = 0.8 mM; [ATZ] = 2.5 mg/L; $t = 60$ min; $T = 25°C$; visible light irradiation	Bismuth molybdate (Bi_2MoO_6) prepared via the hydrothermal method and applied to activate peroxymonosulfate (PMS)	[110]
	Amoxicillin	Ag/TiO_2/mesoporous g-C_3N_4	71	[Catalyst] = 1000 mg L^{-1}; [AMX] = 5 mg L^{-1}; $t = 60$ min	Hospital wastewater, 300 W xenon lamp ($\lambda > 420$ nm) with visible light irradiation	[111]
	Paracetamol	Fe_2O_3-TiO_2	95.85	[Catalyst] = 1.25 g L^{-1}; [PCM] = 30 mg L^{-1}; PH = 11; solar irradiation	Fe_2O_3-TiO_2 is synthesised by the sol-gel method for the degradation of paracetamol in synthetic wastewater	[112]

Table 4: Continued.

Process	Target compound	Materials	Degradation (%)	Conditions	Additional information	References
Ozonation	Bisphenol A	Ni-Fe LDHs/ O_3	100	[BPA] = 10 mg L^{-1}; [catalyst] = 0.3 g L^{-1}; [ozone] = 9.0 mg L^{-1}; [TOC] = 9 mg L^{-1}; [COD] = 32 mg L^{-1}; t = 120 min	Ni-Fe LDH showed effective catalytic performance in the catalytic ozonation of BPA in real secondary effluent wastewater. BPA could be completely removed, and the final removal of TOC and COD was 56% and 68%	[113]
	Atrazine	O_3 (microbubble)	95.3	[ATZ] = 1.16 umol L^{-1}; [ozone] = 1 mg L^{-1}; gas flow: 0.5 L min^{-1}; t = 120 min; T = 20°C	Microbubble ozonation enhanced the degradation of atrazine at different pH levels in a semibatch experiment	[114]
	Amoxicillin	O_3	70	[AMX] = 20 uM; [ozone] = 75 mg L^{-1}; gas flow: 1 L min^{-1}; T = 23°C; PH = 6.8	The degradation of amoxicillin by ozonation resulted in 70% removal with an ozone dose of 75 mg L^{-1}	[115]
	Paracetamol	MgO/O_3	100	Ozone dose: 1.8 mg/min; [MgO] = 0.1 g L^{-1}; [PCM] = 50 mg L^{-1}; t = 15 min; PH = 5.4	MgO powder was used as a catalyst for the ozonation of paracetamol; total degradation and 94% mineralisation were achieved at reaction times of 15 min	[116]
Electrochemical	Bisphenol A	Nb/BDD	90	[BPA] = 5.0 mM; flow rate = 384 mL min^{-1}; j = 42.7 mA cm^{-2}; t = 4 h; PH = [7–10]; T = [6–20°C]	The application of electrochemical oxidation has shown high removal efficiency of BPA	[117]
	Atrazine	Nb/BDD	99	1.5 L of 100 ug L^{-1} atrazine 0.03 M Na_2SO_4; j = 2 mA cm^{-2}; PH = 3; T = 23°C; t = 45 min; batch mode with undivided cylindrical cell	More than 99% of ATZ was removed by anodic oxidation; the atrazine-desethyl-desisopropyl (DEDIA) was the most important by-product recorded	[118]
	Amoxicillin	Ti/Cu-PbO_2	99.4	250 mL of 100 mg L^{-1} amoxicillin; 0.1 M Na_2SO_4; j = 30 mA cm^{-2}; PH 3.5; room temperature; t = 150 min; pseudo-first-order reaction	Copper-doped PbO_2 electrode was prepared and used as an anode to degrade amoxicillin in a laboratory-scale experiment. The optimum removal of AMX and COD was 99.4% and 46.3% after 150 minutes of electrolysis	[119]
	Paracetamol	Pt/Ag-Agcl	90	250 mL of 20 mg L^{-1} paracetamol; 0.1 M Na_2SO_4; j = 5.1 mA cm^{-2}; PH = 4; t = 240 min	The maximum removal of PCM, COD, and TOC reached 90%, 82%, and 65% after 240 min, with the formation of by-products (hydroquinone, benzoquinone, and carboxylic acid) during the electrolysis process	[120]

transfer. Insoluble electrodes (Nb/BDD; Ti/Cu-PbO_2; and Pt/Ag-Agcl) are commonly used [117, 119, 120] to promote the generation of hydroxyl radicals and allow the complete oxidation of a large number of organic molecules contained in wastewater.

AOPs can be used to treat aqueous solutions loaded with organic matter, either as a pretreatment to transform the refractory compounds into biodegradable products or as a final treatment to completely mineralize the organic compounds [106]. They have several advantages in terms of high oxidation efficiency, their ability to treat almost all organic matter, faster reaction rates, and absence of secondary pollution, and they have no negative impact on the environment [141]. The main disadvantage of these processes is

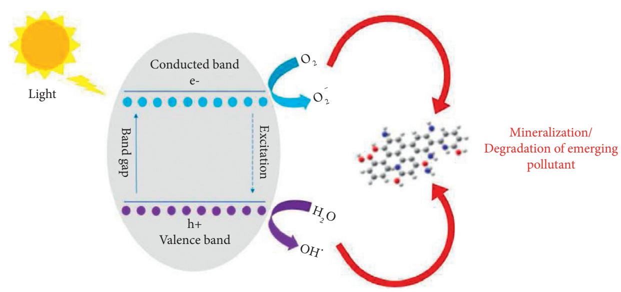


Figure 5: Mechanism of photocatalytic degradation of EPs.

the associated capital and operating costs. Therefore, environment-friendly energy-saving techniques should be used, such as solar energy, for oxidation [140].

6.3. Biological Processes. Biological treatment is a widely used technology for treating wastewater and is also frequently applied for the treatment of emerging pollutants [74]. Many researchers have studied the biodegradation of these substances in various systems, including conventional activated sludge [142], which is the main bioprocess for organic matter removal in wastewater treatment plants [143], sequential batch reactors (SBRs) [144], constructed wetlands (CWs) [145], membrane bioreactors, etc.

In general, the biodegradation of micropollutants in biological processes depends on several factors: the nature of the micropollutants, the characteristics of the organisms, the type of water matrix, and the operating conditions [15, 117]. Table 5 presents biological processes used for the degradation of the studied emerging pollutants. It also summarizes the degradation conditions and removal efficiency. Biodegradation is considered to be one of the current methods for treating a wide range of contaminants. It has many advantages in terms of economy, environment friendliness, and low cost [119,151]. In addition, some persistent substances are able to pass through biological wastewater treatment processes [152] and, therefore, have negative effects on the environment. Under these conditions, adsorption and advanced oxidation processes are possible complementary routes for removing these contaminants.

6.4. Hybrid Processes. The limited effectiveness of conventional treatment processes for the removal of many emerging pollutants is encouraging the development of hybrid technologies using the different removal potential of different processes to overcome the limitations of the removal of these compounds [153]. The hybrid process is based on combining two or more treatment techniques for the effective removal of recalcitrant micropollutants [152].

Advanced oxidation techniques have been used to improve the efficiency of the different physical and biological treatment processes (Table 6). For instance, Jiang et al. [154] studied adsorption treatment in combination with photocatalysis to treat bisphenol A. BPA molecules are rapidly adsorbed onto boron- and nitrogen-codoped graphene aerogels and eventually mineralize after exposure to visible light. Moreover, Taylor et al. [156] suggested a pretreatment in the Fenton process to disintegrate amoxicillin and, thus, facilitate its removal by biodegradation. On the other hand, Iborra-clar et al. [157] investigated the biodegradation of paracetamol in the activated sludge process in combination with activated carbon (AC), and the system was able to degrade paracetamol in wastewater completely.

7. Challenges and Perspectives

Different treatment processes have been developed to limit the release of emerging contaminants into the environment, and each has shown its advantages and disadvantages. Adsorption is a simple and low-energy process, but it requires a large amount of adsorbent. The complete mineralization of pollutants characterizes AOPs under low operating conditions. For instance, photocatalytic water treatment uses sunlight as a nonpolluting energy source, making it one of the most promising methods for the degradation of pollutants. However, photocatalysis has the disadvantage of low light transmittance and slow reaction kinetics [158], limiting its large-scale application. Biological treatment is a widely used method for wastewater treatment, but it is less effective in removing some emerging micropollutants, allowing them to be released into the aquatic environment.

As there is no perfect treatment method, researchers have attempted to provide integrated solutions, such as the coupling of adsorption and photocatalysis, which are simple and environment-friendly processes that appear to be effective in removing micropollutants. This hybrid technology integrates both techniques' advantages through the removal of pollutants from the aqueous phase by adsorption and the degradation of trace organic pollutants by photocatalysis. The main hybrid technologies to be considered are simultaneous combination (one step) and separate coupling (two steps). Figure 6 shows the operation schemes of these technologies.

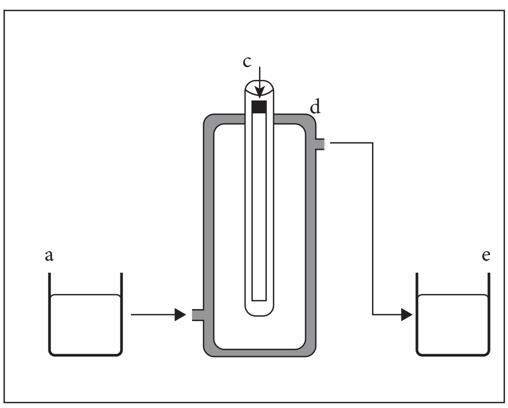
7.1. Simultaneous Coupling of Adsorption and Photocatalysis. Many studies focus on the treatment of different pollutants in water by the combination of adsorption and photocatalysis, as the catalytic reaction is related to surface adsorption. For example, Luo et al. [159] synthesised TiO_2-wood charcoal composites for the removal of bisphenol A and found that synergistic adsorption and photocatalytic degradation were effective in removing hydrophobic bisphenol A. Wang et al. [160] prepared iodine-doped biochar as a photocatalyst adsorbent for the removal of phenol and tetracycline and observed that iodine doping enhances adsorption by creating additional pores and leads to strong photoinduced excitation, which increases the photocatalytic activity of the iodine-doped biochar for the degradation of organic pollutants. Bouyarmane et al. [161] prepared TiO_2-hydroxyapatite nanocomposites for the degradation of drugs in solution under UV light, and the results revealed that the pharmaceuticals were preferentially adsorbed onto the apatite-rich composites, while their photodegradation was more efficient in the TiO_2-rich phases.

Table 5: Biological processes used for the degradation of emerging pollutants.

Pollutant	Process	Type of effluent and operating conditions	Degradation	Additional information	References
Bisphenol A	Aerobic granular sludge	Synthetic municipal wastewater with the following concentrations: COD = 445 mg/L; BPA = 2 mg/L; pH = 7.5–8.0; $T = 20^{\circ}$C; $t = 16$ h	39%	Mixed community of microorganisms in aerobic granular media was used, BPA degraders were active at the beginning of the reactor cycle, and no BPA degradation by-products were detected	[146]
	Sequencing batch biofilm reactor	Synthetic wastewater with the following concentrations: BPA = 10 mg/L; 10 mL of activated sludge; 57.5 g/L of waste iron; PH = 8; $T = 21^{\circ}$C; $t = 100$ min	92%	Acclimatisation of activated sludge with BPA and waste zero-valent iron had a positive effect on BPA removal in a sequential batch biofilm reactor	[144]
Atrazine	Anaerobic moving bed biofilm reactor	Pilot-scale study with synthetic wastewater, ATZ = 0.1 mg/L; COD = 500 mg/L; HRT = 24 h; $T = 32^{\circ}$C; PH = 7.5	ATZ = 60.5% COD = 97.4%	The biofilm moving bed anaerobic reactor showed excellent efficiency in the removal of organic matter and atrazine	[147]
	Denitrifying bioreactors	Laboratory experiments with synthetic wastewater, [ATZ] = 20 ug/L; (NO_3-N) = 1.5 mg/L; $T = 21^{\circ}$C; HRT= (4-8-24–72 h)	ATZ = 53% NO3-N = 100%	NO3-N and atrazine removal increased with increasing HRT. At 4 h, the wood chip bioreactor removed 65% of the NO_3-N and 25% of the atrazine, but at 72 h, the bioreactor removed all NO3-N and 53% of the atrazine	[148]
Amoxicillin	Anaerobic degradation systems	Laboratory experiments with synthetic wastewater; [AMX] = 2500 ug/L; $T = 37^{\circ}$C; PH = 7.2	AMX is completely eliminated	Amoxicillin was completely eliminated under anaerobic conditions. However, analysis identified amoxicillin penicilloic acid, amoxilloic acid, amoxicillin diketopiperazine, and phenol hydroxypyrazine as by-products	[149]
	Anaerobic digestion and aerobic-sequencing batch reactor	Lab-scale combined anaerobic and aerobic processes for swine wastewater treatment containing 19 antibiotics; HRT = 3.3 days; total antibiotic concentrations 99.2 to 339.3 μg/L; COD = 5683 mg/L; [AMX] = 60 ng/L	Antibiotics = 92% COD = 95%	Biodegradation of antibiotics was favoured in the SBR, while the degradation of COD was favoured in the anaerobic reactor	[150]
Paracetamol	Activated sludge	Laboratory experiments with synthetic wastewater; activated sludge comes from an aerobic tank in a wastewater treatment plant. [PCM] = 100 mg/L; $t = 72$ h	99%	The pseudomonas population could eliminate PCM at levels up to 590 mg/L and could also metabolize the PCM-derived metabolites 4-aminophenol, hydroquinone, and 1, 4-benzoquinone at varying levels	[142]
	Constructed wetlands (CWs)	Pilot-scale vertical flow constructed wetland with hospital wastewater; [PCM] = 10 mg/L; HRT = 5 d; media bed: sand and gravel	˃99%	*S. validus* peroxidase enzymes are planted in the CW to control PCM. The vertical flow CW was effective in removing PCM (˃99%) in hospital wastewater treatment	[145]

Table 6: Removal of emerging pollutants by hybrid processes.

Pollutant	Processes	Operating conditions	Removal efficiency	Additional information	References
Bisphenol A	Adsorption - photocatalysis	[BPA] = 20 mg/L; [Photocatalyst] = 1 g/L; $t = 1$ h; $T = 25°C$; batch photoreactor with visible light irradiation	BPA = 96% TOC = 88%	BPA molecules are rapidly adsorbed onto boron and nitrogen codoped graphene aerogels and eventually mineralised upon exposure to visible light	[154]
Atrazine	Adsorption-ozonation	[ATZ] = 0.7 mg/L; [Adsorbent] = 16 mg/L; $[O_3]$ = 19.7 mg/L; $t = 17$ min; $T = 25°C$; batch experiments with synthetic wastewater	ATZ = 90%	A better reduction of atrazine (90%) is obtained when the treatment starts with powdered activated carbon followed by ozone, with a contact time of 17minutes	[155]
Amoxicillin	Fenton-activated sludge	1 mg/L of AMX; 6 mL of H_2O_2 (30% w/w), 4 mL heptahydrated ferrous sulphate ($FeSO_4 \cdot 7H_2O$) solution $T = 40°C$; $t = 70$ min	AMX = 85.13%	The pretreatment in the Fenton process disintegrated the AMX, thus reducing these toxic effects in the subsequent treatment, as the activated sludge can easily degrade the antibiotic	[156]
Paracetamol	Biological-adsorption	2 mg/L of PCM; 1.5 g/L of granular activated carbon	PCM = 100%	The hybrid sequential batch reactor- (SBR-) activated carbon system was able to completely degrade paracetamol in wastewater	[157]



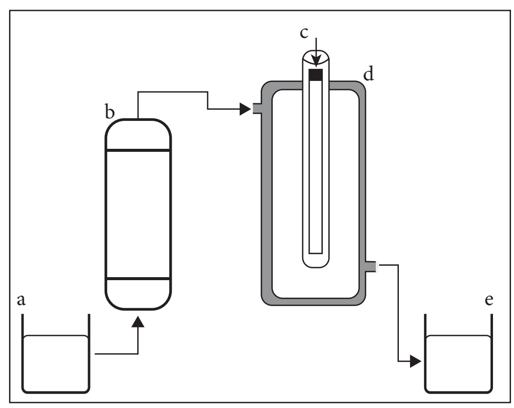


Figure 6: Operation schemes of (a) simultaneous combination (one step) and (b) separate coupling (two steps) of adsorption and photocatalysis (a, wastewater influent; b, adsorption column; c, lamp; d, photocatalytic reactor; and e, wastewater effluent).

7.2. Separate Coupling of Adsorption and Photocatalysis. The literature review shows that several researchers have been interested in the simultaneous combination of adsorption and photocatalysis. However, although this coupling can be carried out simultaneously to obtain the advantages of both techniques in a single step, if problems such as the low use of light and the need for agitation cannot be solved appropriately, the large-scale technical application of photocatalysis appears uncertain. Zhang et al. [158] developed an adsorptive photocatalyst (Zn-doped B_iOI) for the removal of antibiotics from water with a parallel coupling of adsorptive separation followed by photodegradation. The results show that the Zn-doped B_iOI has a removal rate of more than 95% after 5 min of adsorption for the six antibiotics tested. Subsequently, trace contaminants were effectively degraded during the subsequent visible light irradiation process.

Many adsorbent materials have been reported in the literature for their ability to remove different types of emerging pollutants, including carbonaceous materials, agricultural solid waste, and nanomaterials, clays. Some adsorbents are more widely studied than others, such as activated carbon, which is the most frequently used adsorbent for removing EPs. Indeed, much attention should be given to local (Moroccan) materials such as phosphate waste rock, which is generated in large volumes and occupies large areas in mining sites, Moroccan clays, and agricultural waste, by improving their properties to develop effective materials, can be used instead of expensive commercial adsorbents for the removal of micropollutants.

In addition, many photocatalysts (TiO_2, Z_nO, WO_3, etc.) have been studied and found to be suitable for the degradation of emerging pollutants. Among the different photocatalytic semiconductors, titanium dioxide (TiO_2) seems to be the most often applied in water treatment, as it presents better photocatalytic performances. Still, its photoexcitation requires the use of ultraviolet irradiation, which limits the use of solar irradiation. Indeed, the development of new photocatalysts based on low-cost materials and allowing photodegradation under natural irradiation (solar radiation) is a challenge for future work.

8. Conclusions

The widespread presence of emerging pollutants in the environment has attracted worldwide attention because of their severe impacts on the environment and human health. Previous studies have shown that conventional wastewater treatment plants are not effective in treating these contaminants. The data presented in this review summarize the current knowledge on the occurrence, impact, and treatment of bisphenol A, atrazine, amoxicillin, and paracetamol in the environment. These compounds are frequently detected in various aqueous matrices and are among the most common emerging pollutants that can adversely affect humans and the environment. Different treatment methods have been developed to reduce the impacts of these contaminants. Likewise, the adsorption method has the following advantages: reduced energy consumption, simple operating conditions, reduced sludge production, and better adaptation to the removal of environmental pollution from water. Studies have shown that activated carbon is the most widely used adsorbent for the removal of different types of pollutants. In addition, the degradation of contaminants by photocatalysis is a promising method that allows for complete mineralization, without the production of sludge and with scalable applications using sunlight as a renewable and nonpolluting energy source. Titanium dioxide is the most widely applied photocatalyst in water treatment. On this basis, the search for other and more efficient materials is needed. Therefore, the challenge is to develop a treatment process by coupling adsorption and photocatalysis using inexpensive and locally available (Morocco) materials capable of removing/degrading EPs in a wide range of environments rather than being limited to one type of emerging pollutant.

Conflicts of Interest

The authors declare no conflicts of interest.

Acknowledgments

The authors are grateful to the Mohammed VI Polytechnic University of Benguerir and the OCP foundation for supporting this research work.

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