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Review Article

Current status and performance evaluation of emerging advanced remediation techniques for the removal of steroidal hormones in water

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ABSTRACT

Micropollutants such as steroid hormones contaminate water worldwide and cause significant damage. Wastewater treatment plants (WWTPs) partially remove them, and they have low biodegradability leading to their persistence in water bodies. They are endocrine disruptor compounds in aquatic organisms. Various environmental conditions contribute to their persistence in the environment like soil pH, organic matter, soil conditions, and temperature. The aquatic environment is most threatened even when present at very low concentrations. The environment is exposed to steroid hormones from agricultural run-offs, pharmaceutical and industrial waste, and veterinary medicine. Since most WWTPs partially remove contaminants, there is a need for new and improved technologies for higher and more efficient removal of steroid hormones.

This paper discusses the fate and toxicity of steroid hormones in the aquatic environment. It further elucidates the existing and emerging technologies in the treatment of steroid hormones in aquatic environments. Finally, the conclusion of this review along with the current limitations and future research perspectives of hormones present in the environment are elucidated.

1. Introduction

Freshwater is crucially applied for several activities in the household, energy, and industrial sectors worldwide. However, the challenges of wastewater generation from households, and industries into the ecosystem, in addition to excessive usage of limited freshwater, have drastically reduced the quality, and quantity of freshwater resources available to the populace. In meeting up with the ever-increasing demand for freshwater, that's exacerbated by overpopulation, industrialization, and environmental change[1,2]. Thus the production of freshwater from wastewater, and wastewater recycling are the most promising approaches that can solve the freshwater scarcity in society [3,4,5,6,7]. This necessitates the implementation of strategies that can aid in the generation of freshwater, therefore, it is pertinent to consider exploring the potential of recycling steroid hormone (SH) wastewater [8,9,10]. This endeavour is a promising avenue worth exploring.

Steroid hormones (SH) compounds emanate from different activities comprising human excretion, livestock operations, hospital, medical waste system, and industrial discharges [11,12,13,14,15] and are evidenced as major pollutants of the aquatic environment.SH compounds' presence in wastewater treatment plants (WWTPs) at ng.L^{-1} concentration range has adverse effects on both human health and wildlife, thus requiring efficient removal of these hormones from wastewater, before being discharged into the environment or applied for other activities. These SH compound's composition exhibits variance, in terms of geographical location, lifestyle, and physicochemical attributes [16,17,18]. Thus, methods and treatment implementations are not the same. The recycling of SH compounds in wastewater is warranted, to

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combat the overreliance on scarce freshwater resources, and alleviate the discharge of SH compounds into the aquatic ecosystems. Hence, the utilization of recycled SH wastewater could serve as a supplementary means to produce freshwater, which would complement the already existing water reservoirs, particularly in a region plagued by a severe scarcity of freshwater resources.

Given the challenge of SH hormone removal from conventional WWTPs and the exploration of common biological treatment, improved remediation methods are needed to mitigate their detrimental effects on the ecosystem and the public's well-being, given the physicochemical properties of SH compounds [19,20].

Accordingly, the aim of this review thus focuses on (1) the presence of SH compounds in soil, and wastewater, including their concentration ranges, and half-life existence in WWTPs, (2) the issues associated with overall well-being and the ecosystem quality brought on by SHs presence in the environment, (3) the advanced wastewater treatment approaches (physical, biological, and chemical) explored for the elimination of SH compounds, (4) the current scientific evolution and landmark on the use of emerging biogenic nano-architecture materials for the photocatalytic degradation of SH which has not been addressed in any review till date, (5) the effect of monovalent and divalent cation and anion background electrolytes on SH removal efficiency and (6) the limitation of individual treatment approach, and suggested directions for future studies, the combination of two methods, and best treatment technologies in the removal of SH compounds without generation of secondary pollutants. This type of review is necessary at this point because the security of water bodies is one of the most crucial tasks in this 21st century. Moreover, this review will provide updated information for researchers on the current state of development on SH remediation techniques which can serve as guidelines. Furthermore, the study of this review will be beneficial to researchers interested in the development of ideal effective treatment technologies in the removal of SH compounds for wastewater remediation. The knowledge from this review work could also assist readers, researchers, investors, policymakers, and industries that are interested in steroid hormone effluent treatment in their decision-making. In addition, this review work is important for knowledge expansion on the current state-of-the-art research on SH clean-up. Also, the findings of this study can inform global efforts to improve water quality. Finally, the research gaps and future directions identified in this review are important to stimulate further investigation and innovation in the field of water remediation technologies.

2. Review methodology

A systematic review of the literature was carried out by searching several international scientific search engines and repositories like Web of Science, Scopus, Nature, Science Direct, Springer, Wily & Sons, Google Scholar, Taylor & Francis, and PubMed to retrieve relevant articles related to the theme tune of the study. The English keywords utilized in the search include steroid hormones, biogenic materials, nanomaterials, adsorption, toxicity, fate, and removal technologies. These keywords were organized in various combs using the "AND" and "OR" operators. Thereafter, all the co-authors manually screened all the published articles downloaded until January 2024 based on the titles for the elimination of duplicate entries. Additionally, any publications that were missed by internet hunts and perceived to be relevant were manually searched for using the bibliographies. Many papers were found and then sifted according to preset criteria. The first criterion was language, and articles that were not written in English language were not considered at all. Another criterion was perusing the titles and abstracts and expunging the papers that are non-original articles, opinions, encyclopedias, and seminar/conference abstracts. Also, articles where the keywords were not explicitly discoursed or where the full text was not accessible were excluded. In addition, experimental papers not investigating the performance of advanced remediation techniques for the elimination of SH in water were not considered in the core analysis. Thereafter, discrepancies were settled after a cooperative article review and discussion, and publications that met the aforementioned criteria were considered suitable for the review study. Finally, a total of 205 articles were read, analyzed, and discussed for the qualitative synthesis of the current review in a systematic manner. A diagrammatic view of the review methodology is shown in Fig. 1.

3. The fate and toxicity of steroid hormones in the aquatic environment

SHs are naturally occurring hormones that are very important in practically all vertebrates. They regulate several physiological functions including stress responses, reproduction, sexual maturity, and osmoregulation [21]. Furthermore, numerous synthetic steroid hormones have found widespread usage in veterinary and human pharmaceuticals. In addition to improved analytical methodology and increased awareness of their significance, their widespread usage in veterinary and human pharmaceuticals has consequently led to a surge in their detection in aquatic environments [22,23]. Not only has the frequent and accurate determination of both natural and synthetic steroid hormones been aided by significant advances in environmental and analytical chemistry, it has equally been possible to also identify and quantify their transformation products, implying that steroid hormones are widespread aquatic contaminants [24].

The fate of steroid hormones in the marine ecosystem has continued to raise serious concern owing to their propensity to disrupt the endocrine system of marine creatures and consequently upset the dynamic balance of the aquatic ecosystem [25,26]. The presence of steroid hormones in the aquatic environment could emanate from various sources comprising the release of steroid hormones through the faeces and urine of animals and humans, the release of used hormone supplements in dairy cattle, and introduction by agricultural runoff into water bodies, among others [27,28].

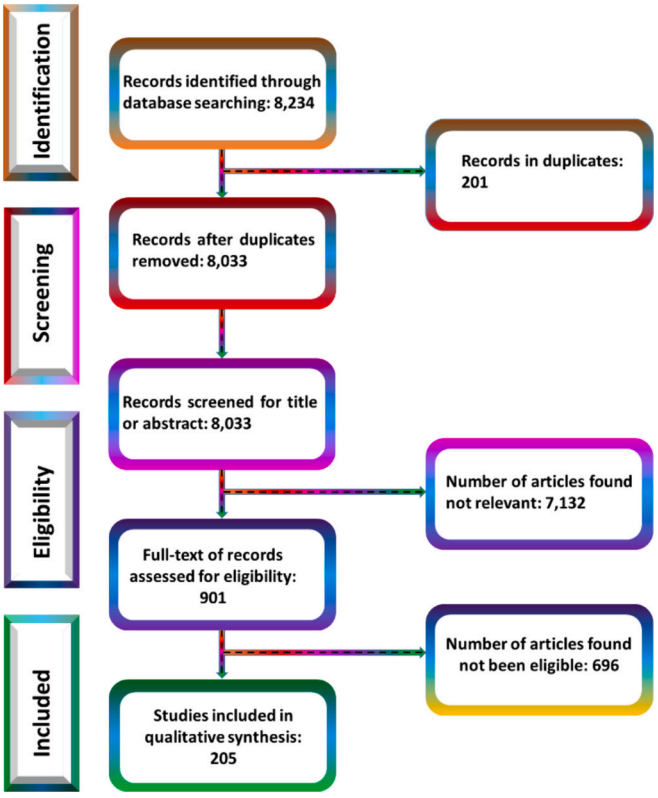

Fig. 1. Diagrammatic representation of the review methodology.

Once these steroid hormones get into the environment, their transport and transformation are regulated or governed by various processes including sorption, biodegradation, photodegradation, and chemical transformation. Steroid hormones have the tendency to be bound to suspended particles in the form of organic matter or sediments in water [29]. The sorption of steroid hormones to these suspended particles implies that they are less likely to be transported away from contaminated sites. This can reduce the mobility of SHs in the aquatic environment. Consequently, the spread of contamination is limited and hence, the potential for exposure to aquatic organisms is significantly reduced. Similarly, the degradation rate of steroid hormones is widely affected by their bioavailability. The sorption of steroid hormones to suspended particles in water indicates that they are less accessible to degrading microorganisms. This results in increased persistence of SHs in the environment [30]. Photodegradation equally affects the transport and transformation of steroid hormones in the environment. The term is used to describe a phenomenon that occurs when a substance is broken down under the influence of light [31,32,33]. The exposure of steroid hormones to light is accompanied by the formation of degradation products [34]. While this culminates in a reduction of the concentration of SHs in the environmental matrix, the newly formed degradation products have properties that are different from those of the parent compounds. For instance, they could be more or less persistent, toxic, and/or mobile [35,36]. Finally, chemical transformation, a phenomenon describing the change of a substance into a new one through chemical reactions, can also affect the transformation of steroid hormones in the aquatic environment. For example, upon the reaction of steroid hormones with chlorine (a disinfectant used in water treatment processes), chlorinated byproducts could be formed. This transformation could significantly alter the properties of steroid hormones including their toxicity, mobility, and solubility [34]. The steroid hormones could be converted into either more or less water-soluble forms and this subsequently affects their transport in the environment [29].

Steroid hormones pose a potential risk of leaching into groundwater. However, the potential for leaching is controlled by soil/sediment particles as well as the characteristics of the chemicals, hydraulic conditions, and the nature of organic matter. As a function of their moderate hydrophobicity, steroid hormones have a propensity to migrate downwards into groundwater, either in the dissolved state or as colloids or particles [28,37].

It has been reported that detectable concentrations of steroid hormones, even concentrations as low as 1 $ng.L^{-1}$ in fish, are widely associated with adverse effects [38]. In some very specific cases, steroid hormones and their homologues have demonstrated their ability to disrupt the endocrine systems, posing severe health effects [39,40]. In addition, the offspring of some altered aquatic organisms have equally shown an impairment in their reproductive ability owing to exposure to steroid hormones [41]. It has been suggested that exogenous steroids adopt several modes of action including a mimicry of the biological actions of endogenous hormones, a blockage of the synthesis of specific hormone target receptors, a blockage of the synthesis of endogenous homologues, and the antagonizing of the effects of endogenous hormones [42,43]. The vulnerability of aquatic organisms to steroid hormones associated with contamination is extremely high. The predominant consequences of exposure to steroid hormones include reproductive, fertility, developmental, morphological, and behavioural disorders [44]. The modification of fish's sexual traits in both their early and late life stages is the main concern connected to hormone exposure in the aquatic environment. By shrinking the size of the testicles, estrogen has been proven to feminize male fish, whilst androgen and progesterone can cause masculinization in female fish [45,46]. As a result, exposure to hormones in the aquatic environment can lead to reproductive issues, such as changes in the gender of offspring, disruption of egg production, decreased spawning rates, and the induction of vitellogenin (VTG) production [47]. From the foregoing, it is obvious that the transport and transformation exhibited by steroid hormones once they enter the marine ecosystem have far-reaching ecological consequences. It is therefore important to protect environmental integrity by engaging several efforts in mitigating the impacts of SHs, especially in the marine ecosystem.

4. Existing technologies for the treatment of steroid hormones

Conventionally, wastewater treatment plants (WWTPs) are the principal sources for contaminant discharge into the hydrological environment due to their poor cleaning processes, which ineffectively eliminate contaminants of rising concern like SH. More specifically, according to chemical fate assessments, increased wastewater treatment levels can sometimes eliminate all SH while also reducing its levels. Besides, because micro-contaminant metabolites or intermediates may still be biologically active, the elimination of SH from effluent does not always imply a decrease in biological activities and seriously affect the ecosystem at large [46,48,49,50,51,52,53]. Studies have shown that the progestins' SH concentration ranges were 0.06–265 $ng.L^{-1}$ [54,55,56–58,59,60,61,62,63,64,65,66,23,67,68,69,70,71,72], 0.2–196.7 $ng.L^{-1}$ for estrogens [73,74,75,52] and <0.12–30.5 $ng.L^{-1}$ for androgens [44,76,77,72] in effluents of WWTPs. The relatively unacceptable concentration is because the majority of WWTPs are not engineered to fully clean micropollutants, particularly when using solely traditional approaches. The physicochemical features of the contaminants and the remediation techniques used affect the cleanup effectiveness of WWTPs [48,49]. Typically, secondary treatment (commonly in activated sludge or membrane biological reactors) is the chief mechanism of contaminant elimination in traditional WWTPs [49]. However, improved cleanup of pollutants may result from the use of extra treatments, often known as tertiary or advanced treatment, in WWTP operations [48,49]. Nonetheless, because scientific studies are always evolving, existing technologies for the additional treatments of SHs are also evolving in a bid to achieve remarkable, eco-efficient, and sustainable SH waste (water) treatment schemes. In addition, the advent of synthetic SH like recalcitrant EE2 has also garnered the morale of scientists to continuously think again about improved techniques for total cleanup of SH in wastewater treatment plants even at very low ($ng.L^{-1}$) concentrations [44,78,52,53]. The existing technologies that have been executed by various researchers for the treatments of SH were discussed in succeeding paragraphs.

4.1. Adsorption

Historically, adsorption is the oldest rapid, popular, eco-economical, and ubiquitous water treatment practice that has been in use for cleaning up water containing SH and the most wildly and oldest cheap functional material used with this technique is activated carbon [79,80,81,82]. Adsorption is an extraction or separation operation [83] in which an adsorbate (part/components of a liquid, gas, or fluid) adhere to the interior and exterior surfaces of an adsorbent (a solid functional material) [84,85,86,87,81,88]. It involves the thermodynamic and kinetic selective interaction of SH compounds from the water phase on adsorbent materials[83]. Parametrically, the mechanistic operation of this technique and its SH removal efficiency is largely governed by temperature, pH, chemical and architectural surface configuration and composition of both the adsorbent and the adsorbate (SH), SH solubility and *pKa*, background electrolytes and additional pollutants or natural organic matter, and ambient and experimental settings [44,86,89,90,91,92,82].

For instance, Esmaeeli [93] used granular AC and powdered AC shown in Fig. 2 to remove 98% estradiol valerate and progesterone in an aqueous solution. However, the removal rapidity of powdered AC is higher than that of granular AC, because of surface-active spots that are within the reach of SHs during the adsorption operation. This observation about powdered AC is consistent with what was reported by Jiang's team [89] for the removal of E2 and EE2 in comparison with other

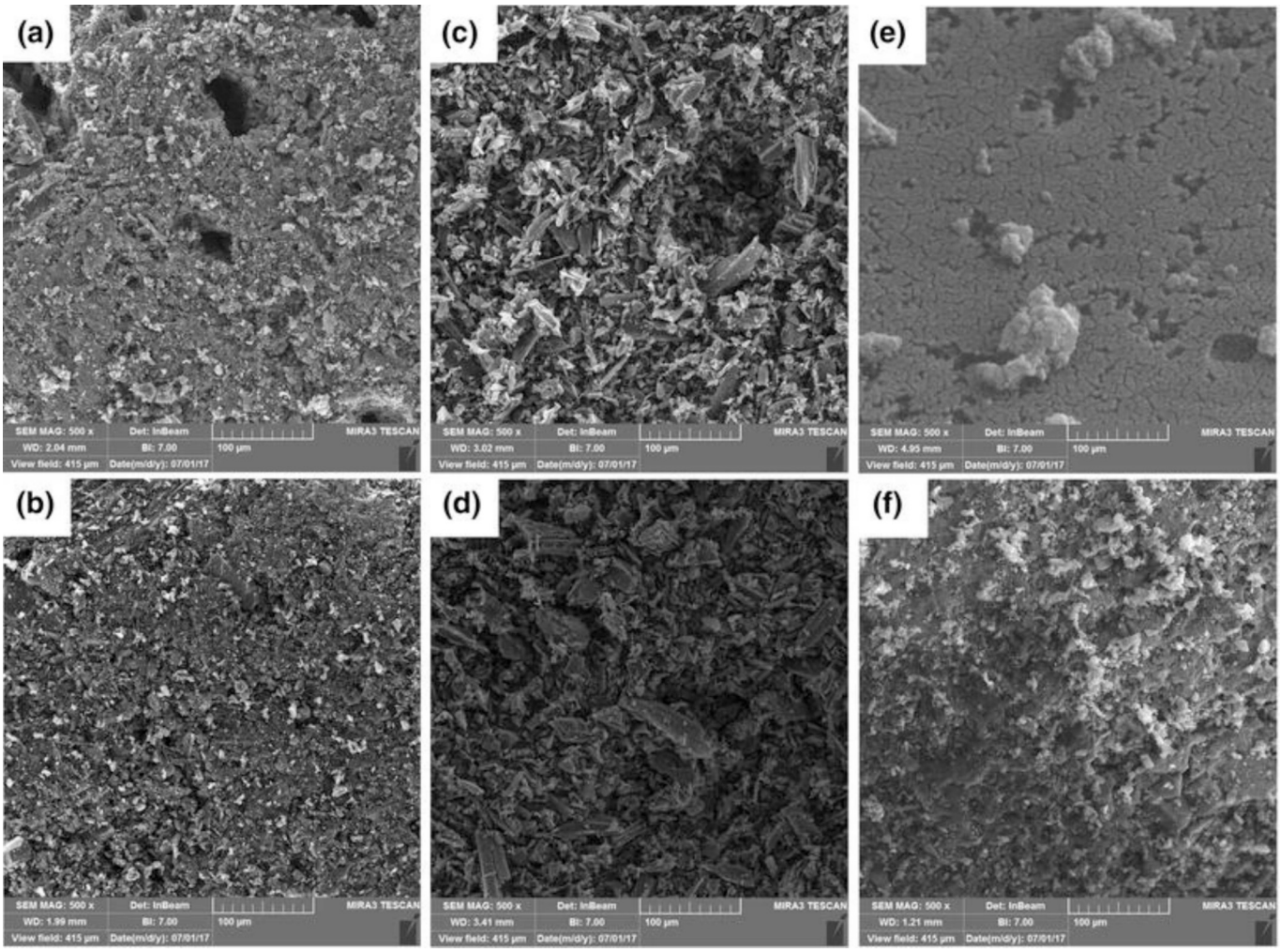

Fig. 2. SEM image of: (a) granular AC prior to adsorption. (b) powdered AC prior to the adsorption. (c) granular AC after removing progesterone. (d) powdered AC after removing progesterone. (e) granular AC after removing estradiol valerate. (f) powdered AC after removing estradiol valerate[93].

materials like rGO, BC, SWCNT, and MWCNT.

In another study, E1, E2, E3, and EE2 from the WWTP sample were adsorbed using Magnetic graphene oxide. In an uptick temperature-dependent manner, the carbon-based adsorbent removed over 90% of each SH except for E3 which showed a downtrend adsorption output with an uptick in temperature [94]. The temperature upsurge adsorption dependency manner was consistent with the one observed for BPA and EE2 using sphere-like AI-based MOF adsorbent shown in Fig. 3 [80]. Similarly, a remarkable 100% removal efficiency was also obtained for the adsorption of E2 from municipal landfill leachate using biochar, spent coffee grounds, almond shells, and potato dextrose agar adsorbent [95]. Some of the remarkable records of over 70% efficiency in the use of the adsorption technique in SH removal from water using various solid materials are presented in Table 1.

4.2. Membrane filtration

The membrane filtration methods shown in Fig. 4 are an auspicious wastewater treatment practice for eliminating different SH micropollutants because they consume less energy [109,110]. Although the UF membrane's big pore size trammels its proficiency in cleaning up SH waters, NF is commonly used owing to its proficient corporeal nature profile of separation and membrane architecture [110]. In a study carried out by [111], it was reported that several NF and RO membranes had their BPA rejections from a test solution of 50 mg.L^{-1} at 10 bar of applied pressure compared. Although the rejection of the loose NF 270 membrane was only about 80%, the polyamide-based dense NF and RO membranes used (NF 90, AD SWRO, BW30, and XLE BWRO) demonstrated excellent functionality with virtually total BPA rejection (98%). Contrarily, the CE BWRO membrane made of cellulose acetate displayed a low and varied rejection for BPA [111]. The kind of exceptional record reported for NF is highly in agreement with the 95% efficiency in 3 h that was achieved by the use of tandem enzymatic (peroxidase and laccase) treatment in an NF membrane paired to recycling mode, for the degradation of BPA [109].

Numerous studies that have shown the proficient removal of SHs by membrane-based filtration techniques are summarized in Table 2.

4.3. Biological treatment

The biological treatment method is a potent and one of the most cost-effective ways for eliminating SH micropollutants during water treatment because this method can treat numerous brands of micropollutants rather than one variety and its working principle involves biodegradation through biological-based systems like biofilms, bioreactors, membrane-based reactor, biofilters and cell-immobilized bioreactor [117,44,118,119]. Notably, there are several types of microbes, algal, and fungi that can break down micropollutants according to published research [117,48,118]. For example, immobilized *Rhodococcus sp.* strain JX-2 was used to remove E2 from sewage and cow muck, and degradation efficiency of 64.4 to 100 was achieved in 7 days [120] and this is in accord with the one reported using ultrasonic-assisted *Pseudomonas*

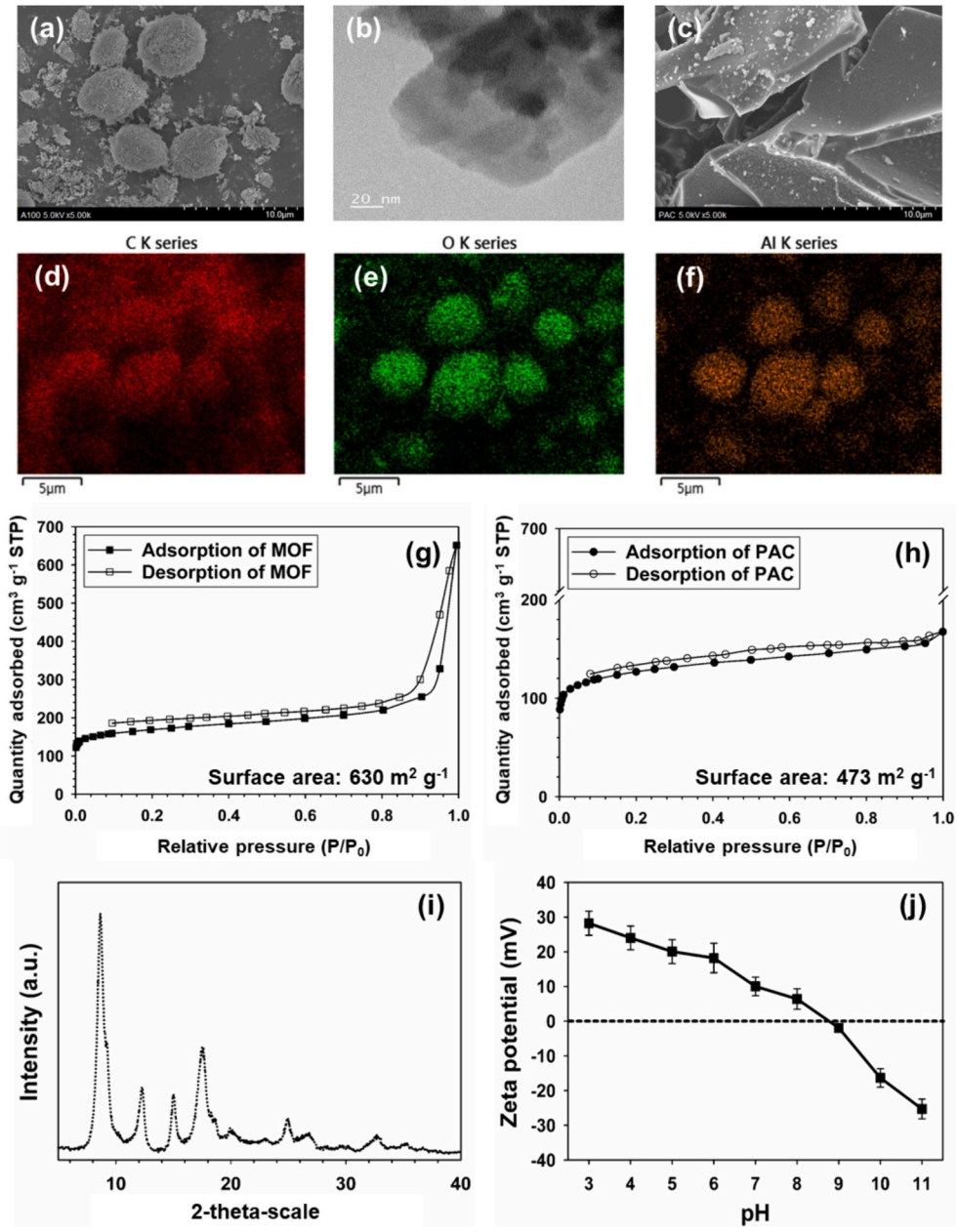

Fig. 3. Analysis of adsorbents by (a) SEM of MOF, (b) TEM of MOF, (c) SEM of PAC, (d-f) SEM-EDS of MOF related to elemental mapping of C, O, and Al, respectively, (g, h) N_2 adsorption-desorption isotherms of PAC and MOF, (i) PXRD of MOF, and (j) zeta potential of MOF[80].

putida which led to the elimination of E1, E2, EE2, BPA, and E3 after 7 days at 100%, 94.86%, 94.90%, 96.56%, and 94.56%, respectively [121]. Conversely, a far superior result was obtained when an immobilized biomass reactor and solar photo-Fenton were employed to biodegrade 95% of BPA present in municipal wastewater in a very short time of just 15 min compared to days reported above to achieve such an excellent output and this is due to synergetic work of adsorption immobilized biomass reactor and photodegradation work by solar photo-Fenton [122]. In a study conducted by Křesinováet al.[49], the degradation of BPA, E3, E1, E2, and EE2 was explored using the white rot fungus *Pleurotusostreatus HK 35*, a popular comestible commercial mushroom grown on farms. >90% of its degrading efficiency was noted to occur within 12 days of normal laboratory settings. Furthermore, it was looked into if recycled mushroom substrates from local farms might be employed in various batch and trickle-bed reactors to eliminate SHs in both real and fortified municipal effluent models. A pilot-scale trickle-bed reactor (PSTBR) was used to evaluate the reactors under different conditions before being used at a wastewater treatment facility. The outcome demonstrated the the ability of the fungus to biodegrade the SHs to a satisfactory degree (roughly 95%). Lastly, a PSTBR was set up at a effluent treatment facility and satisfactorily ran for 240 h. During that time, the PSTBR was able to eliminate over 76 % of SHs that were present in the wastewater. In comparison to the traditional MBBR and ASP, a different form of MBBR reactor that utilizes sponge as its bio-supporter eliminated BPA, E1, E2, E3, E2Ac, and EE2 with over 85% efficiency and is far superior to the efficiency obtained with the traditional MBBR and ASP [123]. Another author [124] compared SHs removal efficacy of autotrophic and heterotrophic kinds of biomass by employing improved nitrifying activated sludge, ammonia-oxidizing bacteria sludge cultured at lab-scale, and traditional activated sludge from a full-scale WWTP. The SHs (BPA, E1, E2, E3, and EE2) worked upon were taken from municipal WWTP effluent. From the result, it was observed that both improved nitrifying activated sludge and ammonia-oxidizing bacteria sludge demonstrated a negligible degrading capacity

Table 1
Summary of adsorption technique performance for treating steroid hormones.

SH pollutant removed	Functional material/ system used	HRE (%)	Time taken (min)	References
Estradiol valerate	Granular AC	98	480	[93]
EE2	MWCNTs	97	840	[96]
E2	Black tea leaves waste	95.75	20–40	[97]
EE2	Black tea leaves waste	95.25	20–40	[97]
E2	Almond shells	100	4320	[95]
E1	Magnetic GO	95.2	30	[94]
E3	Magnetic GO	88.4	30	[94]
EE2	GO sheets	98.46	50	[98]
E2	Sodium dodecyl sulfate-layered double hydroxide	94	20	[90]
Estradiol valerate	Powdered AC	98	480	[93]
BPA	Bio-MOF-1-derived carbon	>90	720	[99]
EE2	SWCNTs	98	840	[96]
E2	Potato dextrose agar	100	4320	[95]
E2	Spent coffee ground	100	4320	[95]
Progesterone	Granular AC	98	480	[93]
Progesterone	Powdered AC	98	480	[93]
BPA and EE2	AI-based MOF	>90	250	[80]
E2	Granular AC	96.98	20–40	[97]
EE2	Granular AC	97.05	20–40	[97]
E2	GO sheets	97.19	50	[98]
E2	Magnetic GO	79.40	30	[94]
EE2	Magnetic GO	93.20	30	[94]
BPA	MgO-AC crystals	>50	<50	[100]
BPA	AC	>30	<50	[100]
E2	cattle bones BC	95.3	–	[101]
BPA	$CuZnFe_2O_4$ BC composite	>95	1440	[102]
BPA	Spanish broom surface modified cellulose fibers	77	60	[103]
E2	Fe_3O_4/GO	>90	–	[104]
E1	Magnetic-bagasse BC	>90	60	[105]
BPA	coconut shell	69	1440	[106]
E1	MWCNTs	83.2	5	[107]
E2	MWCNTs	89.2	5	[107]
EE2	MWCNTs	91.2	5	[107]
BPA	coir pith	72	1440	[106]
BPA	durian peel	70	1440	[106]
E1, E2, EE2 and BPA	*Eucalyptus globulus* wood AC	100	2880	[108]
E3	*Eucalyptus globulus* wood AC	97	2880	[108]

with <14% SHs removal output. Conversely, the heterotrophic fraction of conventional activated sludge eliminates E2 and 78% of E3. In addition, the clean-up of E1, BPA, and EE2 was obstinately propelled biologically with an elimination proficiency spanning from 10 to 46% for E1, 6–34% for EE2, and 10–39% for BPA. In another biological SHs remediation research, a 91.9 to 99.8% removal efficiency was recorded for BPA in 1350 min by the aerated microalgae culture reactor from the membrane runoffs while the non-aerated experiment exhibited elimination ratios of about 80% [125] and this removal efficiency is far superior to the 44% and 32% reported using assembled bio-filter for E1 and BPA respectively [126]. Toro-Vélez *et al* [127] also constructed tropical horizontal sub-surface wetlands with *Heliconia sp.* and *Phragmites sp.* plants. As shown in Table 3**,** the findings of the experiment show that the removal efficiency of *Heliconia*-wetland was 73.3% for BPA, which was more efficient than the *Phragmites*-wetland 70.2% with the unplanted-wetland coming far behind with 62.2% removal efficiency, and this suggests that a tropical plant can contribute to a better performance of constructed wetland for removing BPA.

Numerous microorganisms have had their aerobic catabolism routes for estrogens identified [135]. Microbial degradation of steroid

Table 2
Summary of membrane filtration technique performance for treating steroid hormones.

SH pollutant removed	Functional material/ system used	HRE (%)	Time taken (min)	References
E1, E2, E3, and BPA	UF	3–42	–	[113]
E1, E2, E3, EE2 and BPA	UF	8–46	–	[114]
BPA	polyamide based membranes	≥98	210	[111]
BPA	cellulose acetate membrane	≥98	210	[111]
E1	UF + AS + flocculants	96.0	21*	[115]
E2	UF + AS + flocculants	96.5	21*	[115]
EE2	UF + AS + flocculants	95.0	21*	[115]
BPA	UF + AS + flocculants	95.0	21*	[115]
E1	UF + Freshwater green alga *Nannochloris sp.*	29	7*	[116]
E2	UF + Freshwater green alga *Nannochloris sp.*	60	7*	[116]
EE2	UF + Freshwater green alga *Nannochloris sp.*	60	7*	[116]
BPA	UF + Freshwater green alga *Nannochloris sp.*	46	7*	[116]

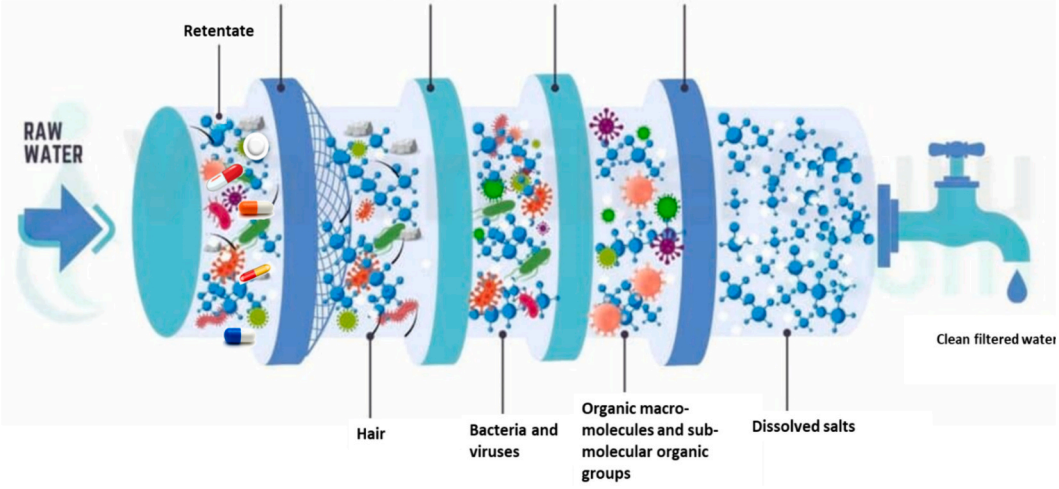

Fig. 4. Membrane filtration techniques [112].

Table 3
Summary of biological treatment technique performance for treating steroid hormones.

SH pollutant removed	Functional material/ system used	HRE (%)	Time taken (min)	References
BPA, E1, E2, E3, and EE2	*Pleurotusostreatus HK 35* bed bioreactor	>90	1440	[49]
BAP	aerated microalgae culture reactor	91.9–99.8	1350	[125]
BPA, E1, E2, E3, and EE2	enriched nitrifying activated sludge	>14	–	[124]
BPA, E1, E2, E3, and EE2	Enriched ammonia-oxidizing bacteria sludge	>14	–	[124]
E1	assembled bio-filter	44	–	[126]
BPA	assembled bio-filter	32	–	[126]
BPA	*Heliconia sp.*	73.3	–	[127]
BPA	*Phragmites sp*	70.2	–	[127]
E2	Conventional AS	100	–	[124]
E3	Conventional AS	78	–	[124]
E1	Conventional AS	10–46	–	[124]
EE2	Conventional AS	6–34	–	[124]
BPA	Conventional AS	10–39	–	[124]
E2	*Rhodococcus sp.* JX-2	64.4–100	7*	[120]
BPA	Immobilized biomass reactor and solar photo-Fenton	95	15	[122]
E1	SBMBBR	89.6	1440	[123]
E2	SBMBBR	96.2	1440	[123]
E3	SBMBBR	92.5	1440	[123]
EE2	SBMBBR	85.2	1440	[123]
E2Ac	SBMBBR	96.8	1440	[123]
BPA	SBMBBR	77.8	1440	[123]
BPA	*Trametes versicolor* membrane bioreactor	100	5*	[128]
E1	*Pseudomonas putida*	100	7*	[121]
E2	*Pseudomonas putida*	94.86	7*	[121]
EE2	*Pseudomonas putida*	94.90	7*	[121]
BPA	*Pseudomonas putida*	96.56	7*	[121]
E3	*Pseudomonas putida*	94.56	7*	[121]
E1	AS	>95	21*	[115]
E2	AS	>96	21*	[115]
EE2	AS	>93	21*	[115]
BPA	AS	87	21*	[115]
E1	moving bed biofilm reactor–membrane bioreactor	>99	–	[129]
E2	moving bed biofilm reactor–membrane bioreactor	>99	–	[129]
E3	moving bed biofilm reactor–membrane bioreactor	>99	–	[129]
BPA	conventional membrane bioreactor	34.5	–	[129]
E3	conventional membrane bioreactor	39.9	–	[129]
BPA	moving bed biofilm reactor–membrane bioreactor	>99	–	[129]
E1	MBR	96	–	[130]
E2	MBR	99	–	[130]
EE2	MBR	93	–	[130]
E3	MBR	97	–	[130]
E2Ac	MBR	99	–	[130]
BPA	MBR	94	–	[130]
E1	MBR-granular AC	100	–	[130]
E2	MBR-granular AC	99	–	[130]
EE2	MBR-granular AC	99	–	[130]
E3	MBR-granular AC	99	–	[130]
E2Ac	MBR-granular AC	100	–	[130]
BPA	MBR-granular AC	100	–	[130]
E1	MBR-powdered AC	99	–	[130]
E2	MBR-powdered AC	99	–	[130]
EE2	MBR-powdered AC	98	–	[130]
E3	MBR-powdered AC	98	–	[130]
E2Ac	MBR-powdered AC	99	–	[130]
BPA	MBR-powdered AC	99	–	[130]
E2	*Selenastrum capricornutum*	88–100	7*	[131]
EE2	*Selenastrum capricornutum*	60–95	7*	[131]
E2	*Chlamydomonas reinhardtii*	100	7*	[131]
EE2	*Chlamydomonas reinhardtii*	76–100	7*	[131]
E2	*Scenedesmus dimorphus*	95	8*	[132]
EE2	*Scenedesmus dimorphus*	85	8*	[132]
EE2	freshwater green alga *Desmodesmus subspicatus*	60	4320	[133]
E2	Moving bed biofilm reactor	84.7	–	[134]

estrogens can either be aerobic or anaerobic. In microbial degradation, several enzymes were used in the breakdown. Bacteria associated with steroid breakdown use IpdAB, a putative CoA transferase (CoT) responsible for the cleavage of a carbon-carbon bond [136]. Oestrogens can be degraded under aerobic conditions *via* the 4,5-seco route in which, in the A-ring, the core ring splits between C-4 and C-5. The ring degradation process involves several coenzyme A (CoA)-esters. Pyridinestrone acid (PEA) and 4-nor-estrogenic acid are products produced by this mechanism and pathway without further products. The 17-oestradiol dehydrogenase oxidizes E2 to E1, After the C-4 of E1 has been hydroxylated by the monooxygenase oestrone 4-hydroxylase enzyme, the catecholic A-ring is subsequently split by meta-cleavage facilitated by the estradiol dioxygenase, 4-hydroxy estrone 4,5-dioxygenase (OecC). In the presence of ammonium, the meta-cleavage result of 4-hydroxyestrone could be through recyclization form the nitrogen-containing product, pyridinestrone acid which is the end product of this mechanism as seen in Fig. 5 ([135,137]). E2 is swiftly converted to E1, also the elimination of E1 is much slower than the conversion of E2 to E1. Conjugates of the estrogens in contact with activated sludge were broken down [137].

Recently, another anaerobic degradation process has been revealed, the Anaerobic degradation 2,3-*seco* pathway. Another degradation method is HIP degradation. Steroid estrogens can also be biodegraded by ligninolytic fungus. Ligninolytic fungi (LF) are fascinating microorganisms that can take part in a wide range of flexible breakdown processes [138]. They can actively remove contaminants with their enzymes. Heme-peroxidases, lignin peroxidase, manganese-dependent peroxidase, and versatile peroxidases are some of the lignin-modifying enzymes (LMEs) that make up the extracellular system and aid in the breakdown process [138]. Ligninolytic enzymes and cytochrome P-450 in ligninolytic fungi can either polymerize the pollutants of interest, in other cases, it can significantly degrade the make-up of the pollutants [138]. Degradation routes by LF can potentially include oxidation at the terminal alkyl chain carbon, carbon-carbon (C—C), and carbon-oxygen (C—O) coupling [138]. Biodegradation of E2 has been reported to have efficiencies of >95 [137], 98 [139], and > 85.5 degradation efficiencies [140].

4.4. Bio-chemical treatment

To clean up contaminated water, biochemical methods including the use of microalgae-based wastewater treatment systems, which use native algae species have been explored. The method is cheap, and the by-products can be used for further applications such as biofuels [141]. The removal of endocrine-disrupting substances (EDCs) using this method has been reported to be successful.

Under repeated loadings of domestic wastewater, the effectiveness of immobilized functional microalgae in E2 removal and degradation has

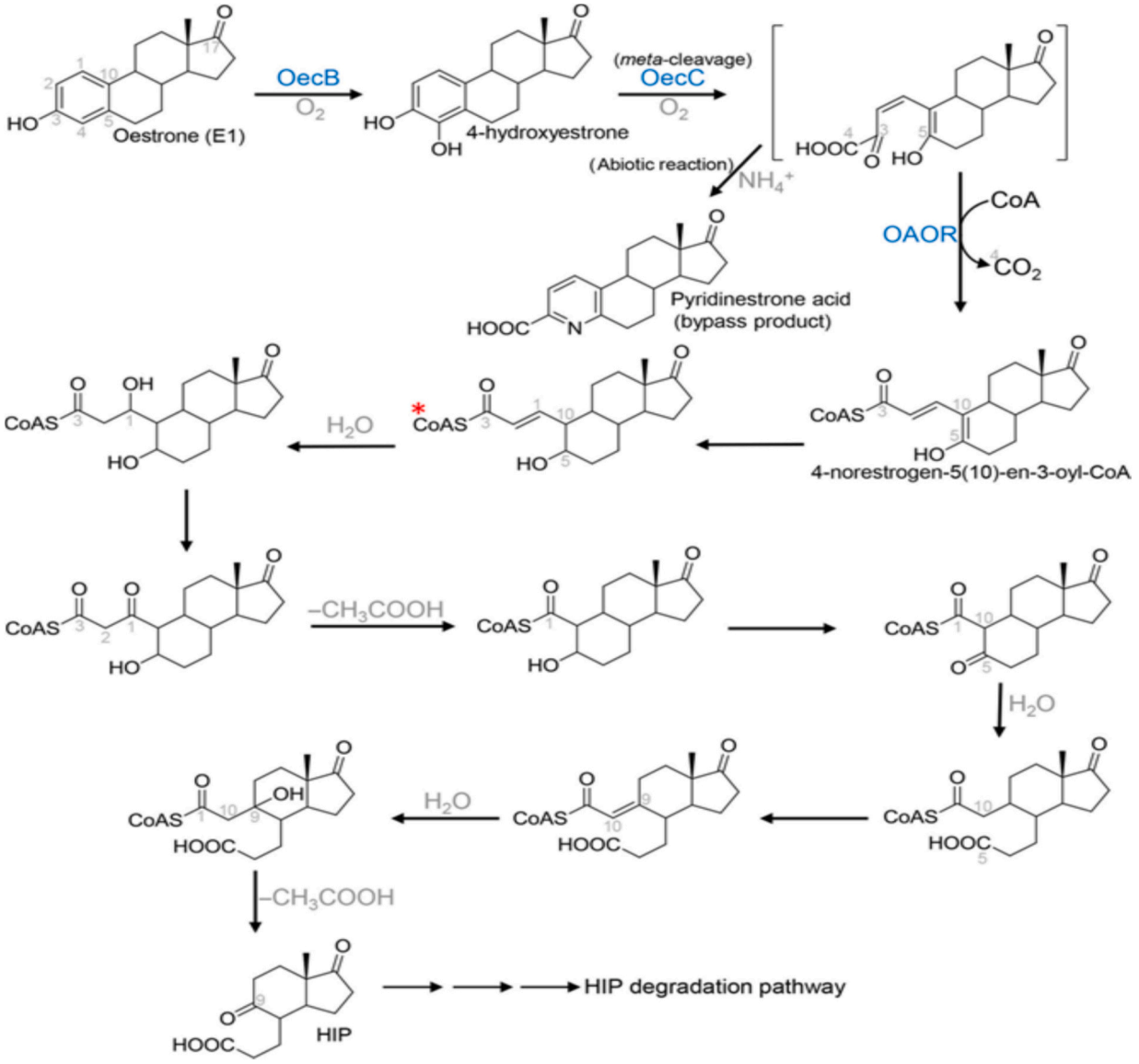

Fig. 5. The bacterial degradation 4,5-*seco* pathway (aerobic) for natural estrogens[135].

been examined (Wang et al., 2020). E2 removal efficiencies of the wastewater treatment employing the immobilized microalgae were significantly improved and were within the ranges of 85–99%. The metabolites also did not contain any other types of natural estrogens, such as estrone or quinone metabolites as shown in Fig. 6, and it was concluded that E2-contaminated wastewater can be recovered using immobilized functional microalgae at over 200 beads mL^{-1}. In this study, the method was concluded to be efficient and secure(Wang et al., 2020).

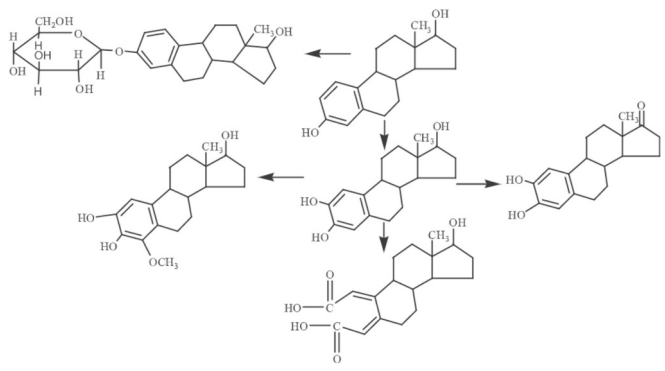

Fig. 6. Pathway for the biotransformation and biodegradation of E2 by the immobilized microalga (Wang et al., 2020).

Interestingly, during ten days of incubation, an 80% removal efficiency of EE2 by free and immobilized microalgae was reported [142]. This study demonstrates that the efficacy of removing EDCs from wastewater effluents is increased when co-immobilized microalgae-based wastewater treatment systems are used [142]. The utilization of microalgae technology is suggested as a feasible alternative for removing EDC from treated wastewater which can also be further improved by immobilizing the microalgae in alginate beads.

A sequencing batch biofilter granular reactor was used to assess the removal of oestrogens such as ethynylestradiol (EE2) from wastewater. It is a brand-new method of biological treatment based on granular aerobic biomass. The lowest average removal percentage was observed with E1 at 64% which might be because E1 can result from the biological treatment of E2, while E2 had an 85% removal percentage[143].

There is still a need for more evaluation of other alternative degradative processes and pathways with lesser metabolites. Intermediate

products should be carefully monitored especially for anaerobic and aerobic processes. Studies should be carried out to adequately understand the details of the transformation process, for each method. This research reflects and encourages the invention of potential alternatives for the removal of oestrogens from water.

4.5. Chemical treatments

Chemical treatment methods have been researched as substitutes to find suitable honing methods to promote the elimination of SHs. This treatment methods include neutralization by pH adjustment or controlling to keep the pH at 7, chemical precipitation such as coagulation, and flocculation using alum, starch, iron materials, activated silica and aluminum salts, and chlorination [117,144,145,146]. In a particular study, the chlorination method was employed for the removal of BPA, the efficiencies of this chemical treatment were reported to be >70% in <30 min. From the same experiment, it was noted that the removal of BPA may be enhanced with an uptick in chlorine dosage, varying pH, and prolonged contact time [147]. Similarly, the elimination of BPA by chlorination was discovered to be around 80% in 20 min as presented in Table 4 [148]. Also, Matamoros and Salvadó [149] employed a tertiary chemical treatment system consisting of coagulation and flocculation followed by lamellar settlement and filtration, disinfection with UV light and chlorination to remove BPA. The removal efficiency was found to be 51%, which is relatively low.

4.6. Advanced oxidation processes

Advanced oxidation processes (AOPs) are an auspicious, sustainable, economical and eco-efficient process that is centered on the *in-situ* production of reactive oxygen species like $^{\cdot}OH$, $HO_2^{\cdot}$, $^{\cdot}O_2^-$, $^1O_2$ and $SO_4^{\cdot-}$ [150] that rapidly initiate the oxidation reaction of SH (usually at the rate of 10^8–10^{11} $M^{-1}.s^{-1}$) to non-selectively attack and mineralize them into a readily biodegradable byproduct that are less toxic or non-toxic molecules like CO_2 and H_2O [151,152,153,154,155,156,157,158,159,160,161,162,20,163,164]. Notably, as shown in Fig. 7, the different available AOPs in wastewater treatment that is often employed to clean up organic pollutants like SH include photocatalytic degradation, Fenton-like oxidation, electrocatalytic oxidation, wet catalytic oxidation, catalytic ozonization and persulphates activation [165,166,167,168,169,154,155,156,158,170,159,141,171,161,172]. AOP technique has attracted the laudable interest of researchers in recent years because it offers superior removal of SH as it does not merely transfer the targeted contaminant from one medium to another, nor lead to the release of toxic byproducts or secondary pollutants as in the case of adsorption [169,173,154,161]. For instance, Sun's research [174] used Fenton oxidation to excellently degrade steroid hormones (E3, DES, E2, BPA, and EE2,) usually found in cow dung. It was remarkably recorded that out of the initial concentration of 97.40,

Table 4
Summary of chemical and biochemical techniques' performance for treating steroid hormones.

SH pollutant removed	Functional material/system used	HRE (%)	Time taken (min)	References
BPA	Chlorination	20	80	[148]
BPA	Coagulation-flocculation followed by lamellar settlement and filtration, disinfection with UV light and chlorination	60–120	51	[149]
BPA	Chlorination	<30	>70	[147]
E1	Flocculation + AS	>96	21*	[115]
E2	Flocculation + AS	>96	21*	[115]
EE2	Flocculation + AS	>94	21*	[115]
BPA	Flocculation + AS	92	21*	[115]

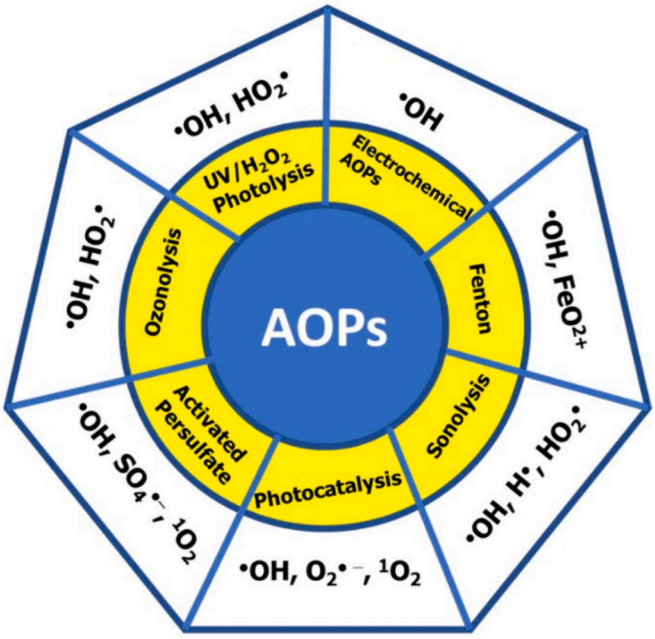

Fig. 7. Different AOPs in effluent treatment and the reactive oxygen species involved [158].

100.22, 95.01, 96.54, and 72.49 $mg.kg^{-1}$ of E3, DES, E2, BPA and EE2, the Fenton oxidation technique was able to clean up 84.9%, 99.1%, 97.8%, 99.5% and 84.5% respectively within 24 h. A similar result was obtained using the same AOP technique with H_2O_2 improved polyacrylonitrile catalyst to degrade over 80% of E1, E2, and EE2 from three different WWTPs in 180 min. In the same spirit, a high degradation efficiency of 97–98% has been reported using Fe(II)-activated peroxy disulphate [162] and glassy carbon granules electrode [151] in an electrochemical AOP technique for the removal of E1, and E2, synthetic BPA, and E2 analogue (EE2) in 120 to 180 min. In addition as per Fenton oxidation for degradation of SHs, it was pointed out by a research group [44] that heterogeneous Fenton's approach type is preferable as it does not lead to sludge generation, unlike homogenous Fenton's style which habitually engenders sludge because of materialization of iron species that are not soluble. As shown in the data gathered in Table 5, it has also been established that employment of UV in Fenton AOP makes a significant reduction in the activation adeptness of the Fenton-like degradation [175].

In another AOP study that is centered on the infusion of one of the most potent oxidants (Ozone (O_3)) into the water, a degradation efficiency of 77%, 99%, 47%, and 96% was achieved for the removal of progesterone, levonorgestrel, medroxyprogesterone, and norethindrone SH respectively [177]. According to a comparative SH degradation research conducted by Si *et al* [114], a combination of the ozonization, AOP method with an ultrafiltration system also gave a good elimination efficacy of E1, E2, E3, EE2, and BPA compared to each method singly. In this experiment, O_3/UF gave a jaw-dropping synergetic removal efficiency of 100%, while ozonization and UF gave a low-grade removal efficiency of 70% and <47% respectively. This very poor removal efficiency of UF alone was ascribed to the fact that the PVDF UF membrane is significantly bigger in terms of pore size compared to SHs. Thus, the PVDF UF membrane finds it difficult to reject SHs. It was further accentuated that the outstanding performance of O_3/UF showed that the joint treatment was far more productive in SH cleanup than independent O_3. More specifically, in the joint process, O_3 performed a more crucial function in the elimination of SHs. This may be for the reason that the UF membrane could only eliminate SHs during the early stages of adsorption, and as soon as the adsorption was saturated, the elimination of SHs largely relied on ozone [114]. The SHs removal efficiency for the O_3 is

Table 5
Summary of performance of advanced oxidation process techniques for the treatment of steroid hormones.

SH pollutant removed	Functional material/ system used	HRE (%)	Time taken (min)	References
E3	UV/Fe (II)/H_2O_2	84.90	1440	[174]
DES	UV/Fe (II)/H_2O_2	99.1	1440	[174]
E2	UV/Fe (II)/H_2O_2	97.8	1440	[174]
BPA	UV/Fe (II)/H_2O_2	99.5	1440	[174]
EE2	UV/Fe (II)/H_2O_2	84.5	1440	[174]
E1, E2, E3, and BPA	O_3	>70	–	[113]
E1	Polyacrylonitrile	>80	180	[176]
E2	Polyacrylonitrile	>80	180	[176]
EE2	Polyacrylonitrile	>80	180	[176]
E1, E2, E3, EE2 and BPA	O_3/UF	100	–	[114]
E1, E2, E3, EE2 and BPA	O_3	>70	–	[114]
BPA	Fe(II)-activated peroxy disulfate	97.00	120	[162]
E1, E2 and EE2	Glassy carbon granules electrode	98.00	180	[151]
Progesterone	O_3	77	–	[177]
Levonorgestrel	O_3	99	–	[177]
Medroxyprogesterone	O_3	47	–	[177]
Norethindrone	O_3	96	–	[177]
EE2	UV/TiO_2/foam concrete	>50	210	[178]
EE2	UV/foam concrete	<50	210	[178]
E1	UVC	97.0	50	[179]
E2	UVC	32.0	50	[179]
EE2	UVC	28.2	50	[179]
E1	UVC/H_2O_2	99.7	50	[179]
E2	UVC/H_2O_2	76.4	50	[179]
EE2	UVC/H_2O_2	77.6	50	[179]
E2	UV/TiO_2	85	60	[180]
Progesterone	Fe/H_2O_2	~60	40	[175]
Progesterone	UV/Fe/H_2O_2	>90	40	[175]
E2	UV/TiO_2	93	5	[181]
EE2	UV/Ag–AgCl/$ZnFe_2O_4$	90	240	[182]
BPA	UV/ TiO_2/ nanodiamond	100	100	[183]
E2	UV/nitrogen modified TiO_2	27	60	[184]
E2	UV/GO-TiO_2	48	60	[184]
E2	CNT	99.1	300	[152]
EE2	CNT	98.4	300	[152]
E2	UV/Aeroxide P25 TiO_2	98	60	[184]
E2	UV/nanocrystalline TiO_2	77	60	[184]
E2	UV–vis/nitrogen-modified TiO_2	23	60	[184]
E2	UV–vis/GO-TiO_2	59	60	[184]
E2	UV–vis/Aeroxide P25 TiO_2	86	60	[184]
E2	UV–vis/nano-crystalline TiO_2	40	60	[184]
BPA	Fe(II)/H_2O_2/UV	100	50	[185]
E1, E2 and EE2	UV/AC/TiO_2	~100	60	[186]
EE2	UV/$CaTiO_3$/WS_2	96	120	[187]
EE2	UV/$ZnFe_2O_4$/Ag/rGO	>80	240	[188]
EE2	UV/$ZnFe_2O_4$/rGO	51	240	[188]
EE2	UV/$ZnFe_2O_4$	25	240	[188]
E1 and EE2	MnO_x (*Pseudomonas putida*$^+$)	100	3000	[189]
BPA	MnO_x (*Desmodesmussp*$^+$)	78	>5*	[190]
EE2	MnO_2 (*Pseudomonas putida* strain MnB1$^+$)	95	120	[191]
E2	Pt and Pd (*Desulfovibrio vulgaris*$^+$)	94	1500	[192]
BPA	MnO_2 (*Escherichia coli* strain$^+$)	96.16	60	[193]
BPA	(*Escherichia coli*$^+$)	>95	60	[194]
E1 and E2	Ag (Orange extract$^+$)	100	–	[195]
EE2	Fe/rGO (Tea extract$^+$)	56.1	180	[196]
EE2	rGO/Fe (Tea extract$^+$)	99.9	–	[197]
E3	Fenton-like sodium peroxydisulfate/ Mn_3O_4(Green tea extract$^+$)	97.5	60	[198]
EE2	Fenton-like sodium peroxydisulfate/ Mn_3O_4 (Green tea extract$^+$) Mn_3O_4 (Green tea extract$^+$)	96.4	60	[198]
BPA	UV/ZnO/SnO_2 (*Viscum 19 album*$^+$)	87.36	120	[199]
BPA	UV/ZnO (*Viscum 19 album*$^+$)	>50	120	[199]
BPA	UV/SnO_2 (*Viscum 19 album*$^+$)	>70	120	[199]
BPA	Fenton-like H_2O_2/ Schwertmannite (*Acidithiobacillus ferrooxidans*$^+$)	100	60	[200]
BPA	UV/ZnO (*Rubus fairholmianus* root extract$^+$)	100	90	[201]
BPA	UV/TiO_2 (*Cassia alata* leaf extract$^+$)	68.41	60	[202]
BPA	Sunlight/ $CdMgFe_2O_4$/ TiO_2 (*M. koenigii* plant extract$^+$)	94	126	[203]

Where AC = Activated carbon, AS = activated sludge, BC = biochar, NP/NPs = Nanoparticle(s), SWCNTs = Single-walled carbon nanotubes, CNTs = carbon nanotubes, MWCNTs = Multi-walled carbon nanotubes, rGO = Reduced graphene oxide, GO = Graphene oxide, MOF = metal organic framework, SH = Steroid hormone, MBR = membrane bioreactor, SBMBBR = sponge-based moving bed bioreactor, HRE = Highest removal efficiency, MBBR = Moving bed biofilm reactor, UF = ultra-filtration, DES = diethylstilbestrol, BPA = Bisphenol A, SWR = seawater reverse osmosis, BWRO = brackish water reverse osmosis, RO = reverse osmosis, NF = nanofiltration, * = time taken in day, $^+$ = biogenic entity/source.

consistent with the one reported by Si *et al* [113] and it was additionally noted that organic matter (NaAH> BSA > NaAg) affects the degradation efficiency.

Ma *et al* also discovered in an experiment that AOP using a UVC/H_2O_2 system successfully degrades different SH with E1 coming to the fore, followed by E2 and EE2. It is noteworthy that the photodegradation of E1, E2, and EE2 surged to 99.7%, 76.4%, and 77.6%, respectively when 15 mg/L H_2O_2 was used with the UVC in <51 min compared to when only UVC was used as it was observed that 97% E1 photodegradation needs <60 min, whereas both E2 and E22, need double of this irradiation time (about 120 min) for such an excellent photodegradation output. Although SH has photosensitive phenolic structures, however, E1 has a much stronger absorption of photons than both E2 and EE2 [179]. In another experiment where TiO_2 NPs were used for photocatalytic degradation of natural E2 SH, an efficiency of 85% was reported in 60 min [180]. However, Alvarez's research group [181] somewhat differs in their observation as it was reported that 93% of E2 SH was photo-catalytically degraded in just 5 min using TiO_2 NPs.

Going forward, the advent of biosynthesized NPs through the help of phytochemicals (flavonoids, phenolics, polysaccharides, alkaloids, *etc*) present in various biogenic entities [4,204,205,206,207] has brought remarkable advances to the removal of SHs as it has low toxicity compared to chemically synthesized NPS and also offers complete mineralization of SH pollutants [189,197,198]. For instance, 100% removal of E1 and EE2 was achieved by Furgal's team [189] using biogenic MnO_xNPs that were biofabricated using *Pseudomonas putida.* A

similar jaw-dropping removal efficiency of 95% and 96.16% was also recorded for EE2 and BPA using MnO_2 NPs (Fig. 8**)** biosynthesized using *Pseudomonas putida* strain MnB1 [191] and *Escherichia coli* strain [193] respectively.

The Author [191] based on further investigation confirmed that the presence of background electrolytes and heavy metals reduces the removal efficiency of EE2 slightly. However, it was accentuated that divalent ions, such as Ca^{2+} from $CaCl_2$, have a stronger impact on the detoxification of EE2 by biosynthesized MnO_2 NPs than monovalent ions, such as Na^+ from NaCl because their interaction with the surface sites of MnO_2 NPs is higher. This impact of background electrolytes was in agreement with the report of Liu's team [197] that pointed out that the presence of co-existing anions such as Cl^-, SO_4^{2-} and NO_3^- considerably diminished the removal efficiency of EE2.

Notably, it could be the presence of these background electrolytes and heavy metals in real-life scenario wastewater that made biogenic Fe/rGO deliver a somewhat low EE2 removal efficiency of 41.5% [196] in domestic wastewater. The SEM image of the biogenic Fe/rGO before and after removing EE2 is displayed in Fig. 9**.** In another study, biogenic platinum NPs were found to significantly diminish the estrogenic activity of E2 after delivering an excellent removal efficiency of 94%, and this indicated that a less harmful effluent was generated using this biosynthesized nanocatalyst [192]. This remarkable efficiency is consistent with the 96.4% and 97.5% recorded for EE2 and E3 respectively when Mn_3O_4 NPs biosynthesized using green tea extract was employed [198].

Yang's research group [195] also reported the complete removal of E1 and E2 *via* a Fenton-like oxidation approach using spherical(Fig. 10) Ag NPs bio-fabricated using an orange extract. This outstanding removal efficiency is in parallel with the one (100%) obtained for the degradation of BPA when Schwertmannite shown in **Fig. 11** [200] and ZnO shown in Fig. 12 [201] biosynthesized using *Acidithiobacillus ferrooxidans* and *Rubus fairholmianus* root extract was employed as a biogenic nanocatalyst. Furthermore, it was established that the spent biogenic silver NPs can be efficaciously reused for up to four rounds with elimination efficiencies of 46.0 and 58.3% for E1 and E2 correspondingly and this implies that biosynthesized NPs in AOP technique could be used to proficiently eliminate SHs from both metropolitan wastewater and river water [195]. Table 6, summarized the advantages and disadvantages of the different treatment techniques and more reports on the use of AOP techniques and biogenic NPs for SHs remediation are summarized in Table 5**.**

5. Cost comparison of steroid hormone removal techniques

Without gainsaying, cost analysis is very common in decision-making and it is the focal point of interest for investors, engineers, industries, and policymakers especially when selecting a sustainable treatment technique for eliminating pollutants [242] like steroid hormones. Notably, there has been a dearth of research on techno-economic analysis of various methods for steroid hormone removal. However, based on the very little literature available, it has been reported that most chemical techniques including Fenton oxidation, electrochemical degradation, photocatalytic degradation, sonochemical degradation, and ozonation are more expensive than biological and physical techniques (especially adsorption and ion-exchange) [222] for steroid hormone removal.

Specifically, adsorption is one of the oldest and cheapest methods [243] for the removal of steroid hormones. This is due to the fact that the method involves the use of inexpensive materials like hydrochar, biochar, and activated carbon that can be made from readily available biomass wastes [79,80,81,82]. For example, the estimated cost of manufacturing AC from gasification waste was 0.23 $/kg, which is far cheaper than the activated carbon market price and this can be ascribed to the availability of low-cost wood chips[244]. In another study, the estimated cost for synthesis of distillers grains-derived clay hydrochars, Rice husk biochar, and AC from Sugarcane Bagasse was around ~28 $/ton, ~0.54$/kg and ~ 2.1$/kg which is much less than the present average market price (~688$/ton) of commercial AC, biochar [245,242,246] and popular TiO2 nanophotocatalyst that can cost close to 1000$/kg. Also, biological treatment methods of steroid hormone effluent including adsorption by microbial biomass, algae, and enzyme degradation are also becoming more cost-effective [222] but are still behind adsorption by cheap materials like biomass waste-derived activated carbon, biochar, and hydrochar. In addition, the high energy/electricity cost associated with chemical methods like photocatalytic and sonochemical degradation is another somewhat hindrance to their full-fledged acceptance. Specifically, the efficiency of photocatalysis using nanocatalysts is hooked on the light source, and thus, high energy costs associated with UV light sources can result in less overall

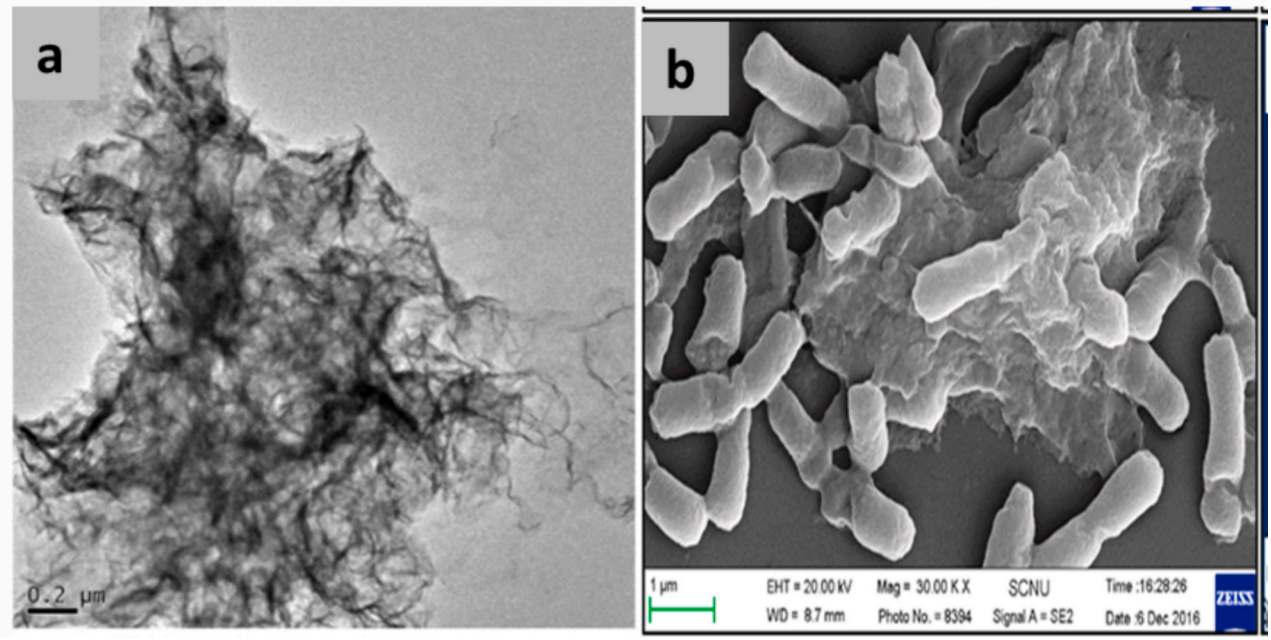

Fig. 8. (a) TEM image of MnO_2 NP produced by *P. putida*[191] (b) SEM image of MnO_2 NP produced by *Escherichia coli* strain[193].

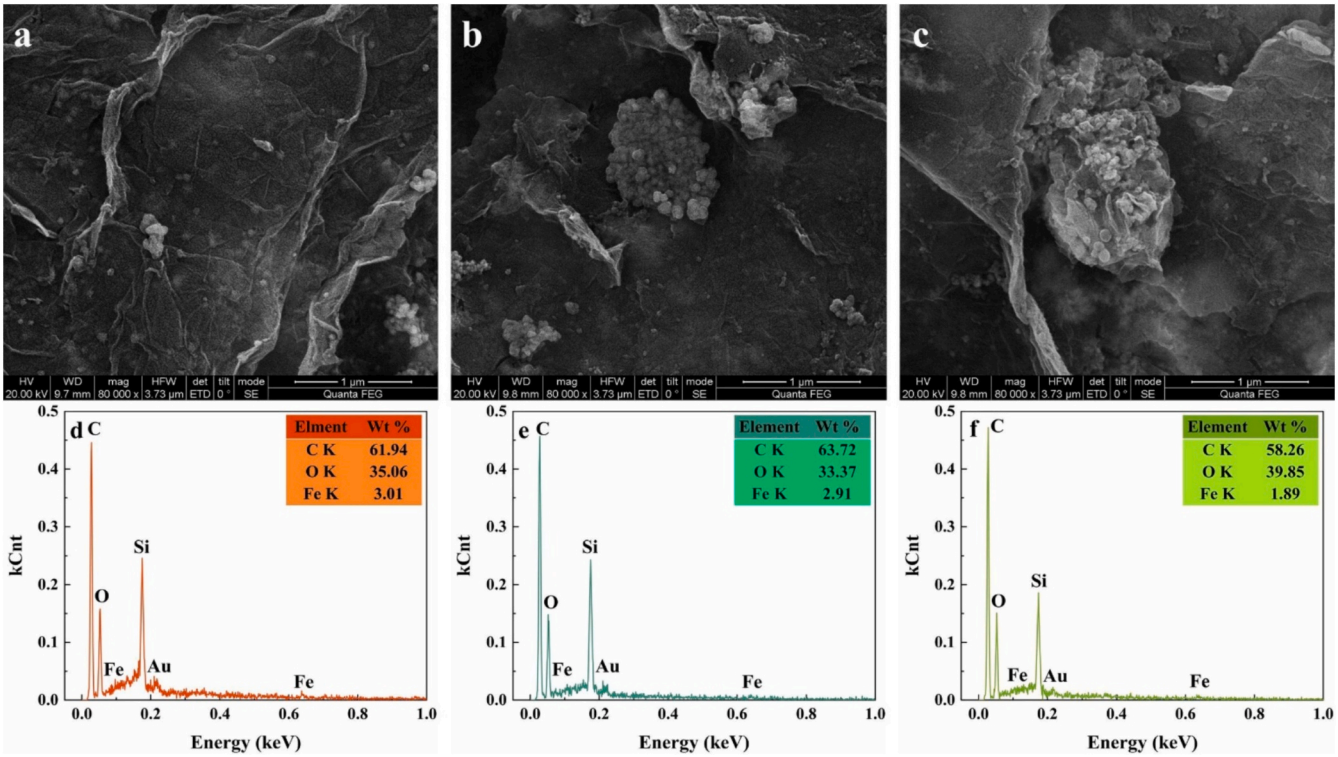

Fig. 9. SEM images and EDS of FeNPs/rGO prior to (a and d), after adsorption (b and e), and after adsorption and Fenton-like oxidation (c and f) [196].

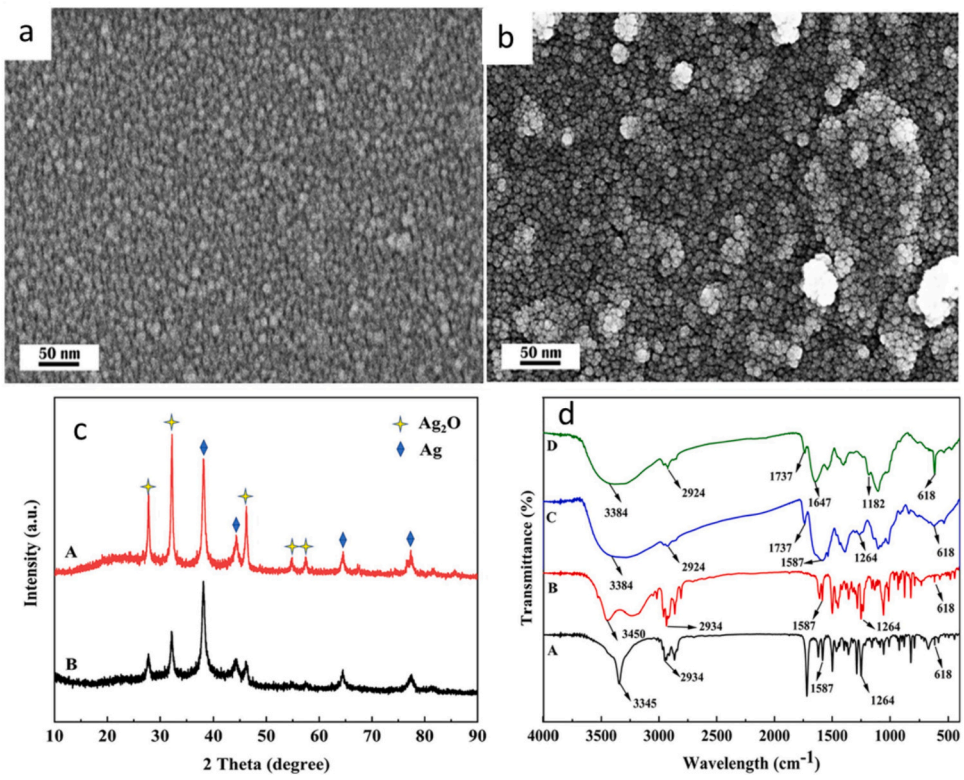

Fig. 10. SEM image (a). Ag NPs-fresh; (b). Ag NPs-reacted; (c). XRD pattern (A. Ag NPs-fresh; B. Ag NPs reacted); (d). FTIR spectra. (A. E_1; B. β-E_2; C. Ag NPs-fresh; D. Ag NPs-reacted) [195].

superiority of process efficiency[247] compared to other methods like adsorption and biological treatment. More specifically, the steroid hormone effluent treatment method like the electrochemical method whose reactor is reliant on high electrical energy[248] is unfavorable for countries faced with energy insecurity, especially in African regions because this will inflict additional operational costs on the process.

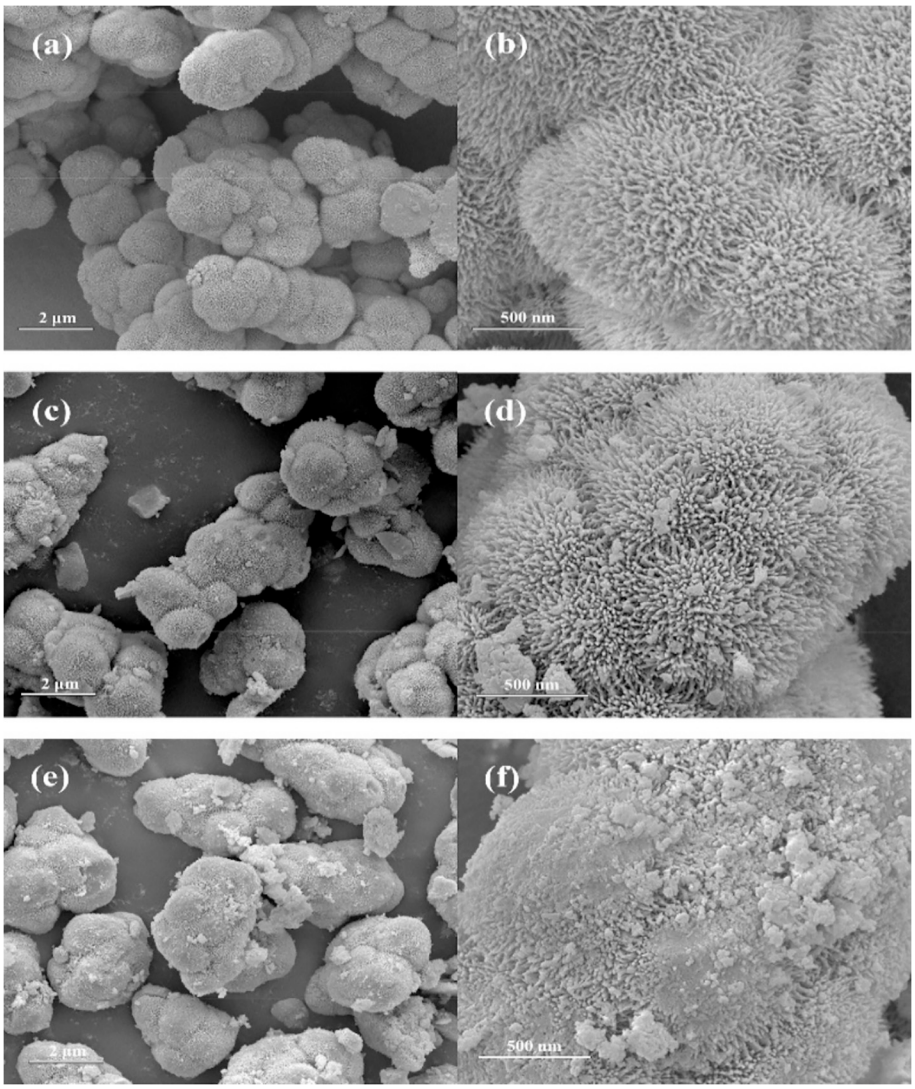

Fig. 11. (a and b) SEM images of the as-fabricated Schwertmannite and the Schwertmannite after being used in (c and d) ultrasonication/H_2O_2 system and (e and f) H_2O_2 system [200].

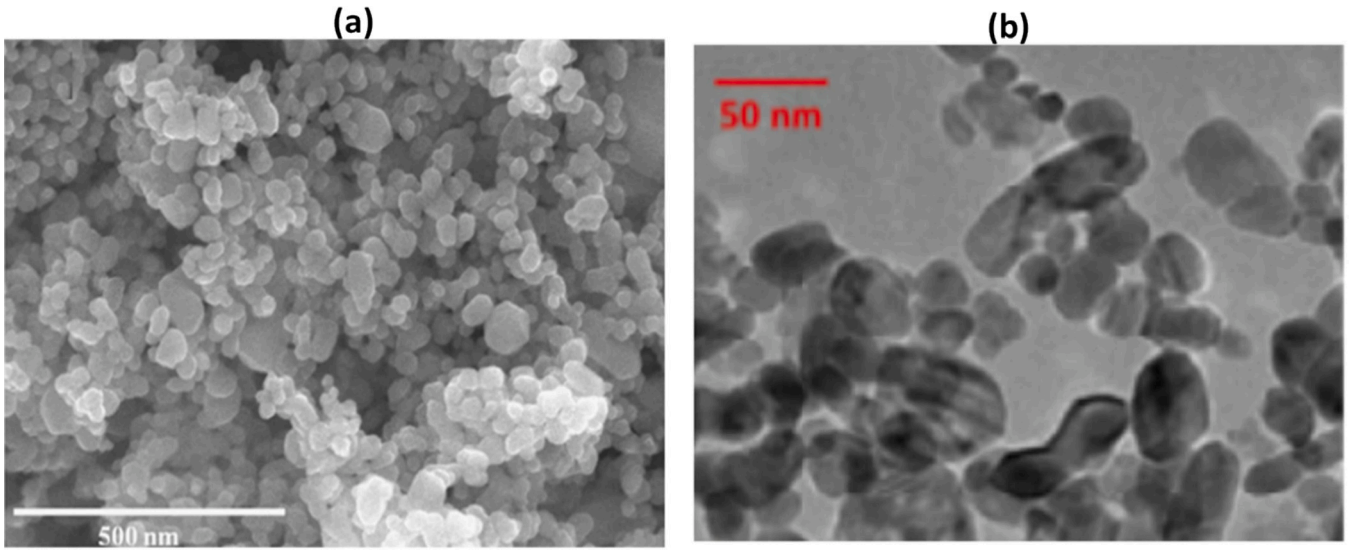

Fig. 12. (a) SEM image and (b) HR-TEM image of biogenic ZnO NPs[201].

Table 6
A Table showing the advantages and disadvantages of different treatment techniques.

Techniques	Advantages	Disadvantages	References
Physical techniques			
Adsorption	Effective elimination of a large range of targeted SH Minimal running costs, suitability for both batch and continuous processes. Simple operation and flexibility. Environmentally friendly method Regeneration is possible and no generation of secondary by-products	Poor adsorbent selectivity Adsorbent disposal complications	([208]; [209]; [210]; [12]; [211]; [212]; [213]; [214]; [215])
Membrane filtration	Environmentally friendly, energy-efficient, and effective in recovering and reusing water without the need for chemical additions Saves energy consumption. No phase change is involved	Concentrated sludge generation Decline in flux A laborious procedure Filter clogging might happen. Expensive start-up costs. Simple fouling of membranes	([216]; [217]; [218]; [219])
Ion exchange	Regeneration: no adsorbent loss It is possible to get zero hardness. Quick procedure of separation Small space needed	Not effective for all SH Pretreatment is necessary for most SH effluents. Ionic competition Matrix fouling Extremely sensitive to the pH of the solution	([220]; [221]; [213])
Nanofiltration and ultra-filtration	Able to eliminate any kind of SH	Expensive and must be used with other methods. High use of pressure and energy. SH molecules continuously obstruct membrane pores. Short lifespan.	([211]; [212]; [222]; [223])
Sedimentation	No energy is needed. Outstanding replicability	Selective process, Ineffective, Lacks accuracy	([213]; [215])
Reverse osmosis	Offer almost complete elimination. Produces pure, and clean water.	Expensive. Requires high pressure and energy consumption. Fouling issues	([222]; [224]; [214]; [223])
Chemical techniques			
Oxidative process	Application simplicity Able to completely break down pollutants within a quick response time	(H_2O) agent needs to be activated by some means. pH dependent. A catalyst is necessary for effective pollutant removal.	([222]; [225])
Ultrasonication and Sonolysis	Simple to use, safe, clean, and energy-efficient without generating any secondary pollutants. Can remove SH at low concentration.	Extreme conditions are required.	([212]; [226]; [218]; [215])
Fenton oxidation	Large SH can be broken down into smaller molecules by strong oxidizing species. Simple to use. cost-friendly reagents	High production of iron sludge. Extended response time. Limitations of functioning at narrow pH range.	([218]; [222]; [195]; [215])
Chemical Precipitation, Coagulation and Flocculation	Good removal of SH and other dissolved organic compounds. little financial outlay Simple process management Minimal running costs	Problem with high sludge output Only fit for a few numbers of SH. Incomplete elimination of SH. Generates sludge in big quantities. The challenge of getting rid of sludge.	([227]; [5]; [205]; [228]; [213]; [229]; [219])
Ozonation	Ozone is a gas that may be applied and does not add to the amount of sludge or wastewater. Effective for SH removal No need to change the temperature or pH Greater germicidal action without the need for chemicals	Short half-life (20 min) demanding continuous application. Fire risks and toxicity problems might arise from ozone generation. Because ozone is less soluble, certain mixing methods are needed. Somewhat more costly than other techniques Generates hazardous byproducts. Less stable. Can not reduce estrone to levels that are not detectable.	([5]; [205]; [221]; [213]; [215])
Ultraviolet irradiation	No sludge generation. Weakens foul odours	Energy depletion. Expensive Limited treatment times. The total degradation of SH may not be achieved by using UV irradiation as the only kind of treatment.	([222]; [230])
Photocatalysis	Foul smells are significantly minimized, and no sludge is formed. mild working conditions that effectively remove color and COD and may be powered by sunlight. Not toxic, relatively low cost, lack of mass transfer limitations, chemically stable, and possible operation at ambient temperatures.	Formation of by-products	([150]; [231]; [232]; [233]; [218]; [234]; [215])
Electrochemical degradation	No chemical consumption and no accumulation of	High flow rates cause SH removal to be less successful	([216]; [217]; [233];

(continued on next page)

Table 6 (*continued*)

Techniques	Advantages	Disadvantages	References
	sludge Generally appropriate for the removal of soluble and less soluble SH Less laborious	than with other approaches. Exorbitant power bills By production of hazardous materials	[235]; [219])
Biological techniques			
Anaerobic	Generation of biogas that may be used again to generate electricity and heat	Longer acclimatization phase High initial investment Odor trouble	([236]; [218]; [213]; [237])
Adsorption by living/dead microbial biomass	Certain SH have a particular affinity for binding with microbial species	Not suitable for many SH	([218]; [222]; [238])
Aerobic	Color removal is facilitating with COD reduction	Microorganisms might become inactive due to co-existence of heavy metals in SH effluent. Pricey Maintenance issue	([239]; [218]; [213])
Enzymatic treatment	Able to efficiently remove aromatic SH that can impede downstream treatment process. Reusable	Tricky procedure for extracting, isolating and purifying enzymes	([240]; [218]; [222]; [214]; [241])

Although, membrane filtration (including the nanofiltration technique) operational cost is less expensive compared to chemical methods, however, it is more costly than physical treatment methods like adsorption[222]. Summarily, the physical treatment method is less costly compared to the biological and membrane treatment methods which are less costly compared to the chemical methods that require high electrical energy for operation.

6. Patents

Although the scaling up of various SH removal techniques discussed in the preceding section is still a challenge. However, a few patents for some of these techniques are available and summarily discussed in this section and summarized in Table 7**.** For example, in 2021, a group of researchers [249] in China got a patent for the use of an ultrasonication method for the recovery of SHs from sediments. As shown in Fig. 13**,** it was revealed that by using the ultrasonic techniques, a recovery of the variety of SHs each can be up to 73% or even more with a maximum recovery that can be up to 100%. This jaw-dropping output is similar to the one record for a similar patent[250] method using silica particles coated with β-CD (β-cyclodextrin). In this method, the β-CD is bounded to the silica particle through at least one crosslinking agent and/or at least one copolymer. The silica@β-CD was then employed in an ultrasonic-assisted method to remove etonorgestrel, BPA, trimegestrone, E1, 17α-dihydroequilin, P4, medrogestone, GSD, norgestrel, 17α-E2, EE2, equilin, E2, testosterone, E3, DSG and E1 from a flowing aqueous phase with over 90% efficiency as shown in Fig. 14 at 15 g/L adsorbent dose. Tremblay and Collias [251] also patent an adsorption method for the removal of SHs. The process involves applying a filter made of AC fibers (ACF) to the liquid SHs. The filter's bulk density ranges from approximately 0.15 gcc to approximately 0.8 g/cm^3. The ACF have diameters between approximately 5 μm and 50 μm and lengths between approximately 10 μm and 10 mm. The ACF remove at least 90% of the SHs at a flow rate of 1 L/min., with a total influent volume often liters at a concentration of 100 nM.

In another patent study[253], a phyto-mediated wastewater treatment bioreactor was developed for treating wastewater effluent containing SHs. The bioreactor was reported to clean up SHs successfully. This might be attributed to the ability of plants to adsorb substances *via* their root and other organs. Also, in 2008, another group of inventors [254] developed effective ultrasound irradiation to degrade and decontaminate SHs in aqueous media. As shown in Fig. 15**,** a degradation efficiency of about 90% was achieved in 25 min for E2, equilin, 17α-E2, E1, 17α-dihydroequilin, EE2, GSD, and LNG. In water and wastewater systems, such as groundwater, surface water, raw drinking water, municipal wastewater, and industrial wastewater (hospital, pharmaceutical), the technique can be applied to break down and remove SHs. The method's inventor accentuated that it does not generate waste sludge or off-gases and doesn't require any chemical additions, pH adjustments, or filtering. This is a significant benefit over procedures like those that employ ozone, which call for removing any leftover ozone from the treated water and destroying any wasted ozone in the off-gas. Additionally, the approach may be used in many environmental matrices and work locations and is not restricted by the properties of water and wastewater, like color, turbidity, or suspended matter [254].

7. Regulatory issues

Regulatory issues surrounding the application of various SHs wastewater treatment technologies typically revolve around concerns related to human health, ecological impact, and compliance with existing water quality guidelines. Thus, the use of various technologies and materials in SH remediation may necessitate adherence to certain regulations and standards, which can differ by nation and region. However, developing reliable and clear guidelines for the use of these technologies can be a challenge. Some specific regulatory considerations include:

Table 7
Available patent technologies for SHs clean-up.

Year	Technique	SHs	Maximum % removal	Patent Number	References (Authors)
2003	Adsorption using activated carbon fibers	Estrogen	90	US6660166B2	[251]
2021	Ultrasonication	E1, testosterone, A4, E2, methyltestosterone, EE, LNG, and PA	100	US20210163525A1	[249]
2017	Ultrasonication using silica particles coated with β-cyclodextrin	Etonorgestrel, BPA, trimegestrone, E1, 17α-dihydroequilin, P4, medrogestone, GSD, norgestrel, 17α-E2, EE2, equilin, E2, testosterone, E3, DSG and E1	>80	US9828458B2	[250]
2011	solid-phase extraction using entrapped β-CD	SHs	>70	US7951890B2	[252]
2022	Phyto-mediated wastewater treatment bioreactor	SHs in effluent mixture	90	US20220274856A1	[253]
2008	ultrasound irradiation	E2, equilin, 17α-E2, E1, 17αdihydroequilin, EE2, GSD, and LNG	74–82	US20080076954A1	[254]
2012	Enzymatic degradation	E2, E1, and E3	80	US8163534B2	[255]
2018	Ion exchange using resin	mixture of estrogens	82.90	US10143938B2	[256]

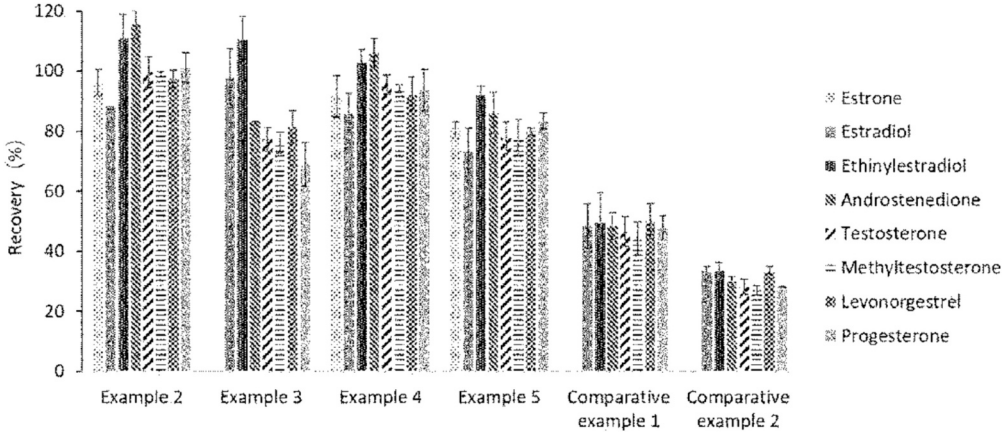

Fig. 13. Percentage removal of SHs using ultrasonic method[249].

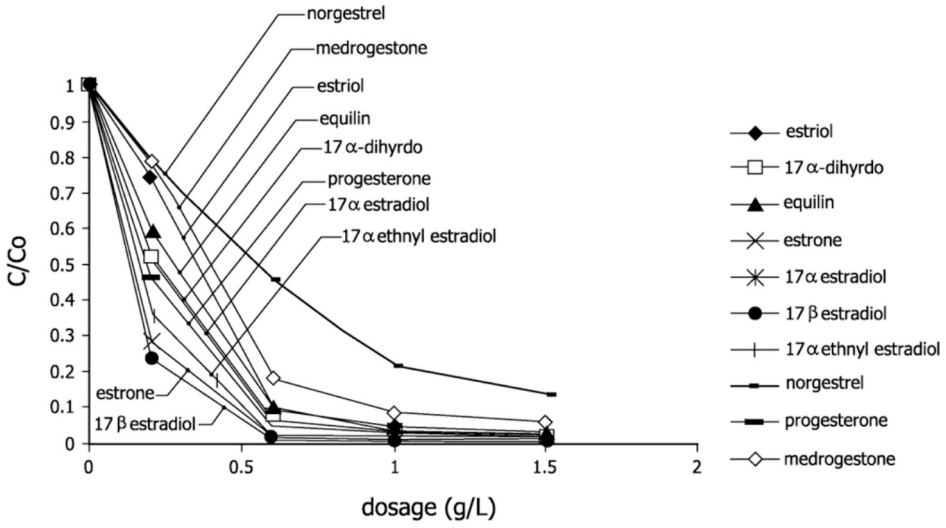

Fig. 14. Percentage removal of SHs using silica particle coated with β-cyclodextrin[250].

- Permitting and Compliance: Industries using SHs wastewater treatment technology might have to apply for permits from regulatory bodies and prove they are following effluent rules and discharge limitations. Requirements for compliance reporting and monitoring might also be enforced.
- Technology Approval: Regulatory bodies may demand validation to guarantee that treatment systems are successful in eliminating steroid hormones from wastewater. This might entail certification and testing procedures to prove adherence to legal requirements.
- Public Health and Safety: Regulatory decisions regarding SHs clean-up technologies may also consider bigger public health and safety concerns, including community perceptions and acceptance of treatment technology.
- Health and Environmental Impact: Regulatory bodies could be worried about the possible health consequences of steroid hormones in the water supply, such as endocrine disruption, impacts on aquatic creatures' ability to reproduce, and possible effects on human health through contact or ingestion of water. Additionally, concerns about latent health and environmental risks linked to some materials (*e.g.* biochar derived from sewage sludge)[257] used with these various techniques can bring about stricter regulations, restraining its possible use as SHs cleaner in wastewater. Specifically for the safe and sustainable manufacturing and utilization of biochar obtained from sewage sludge in adsorption technique, it is imperative to set clear regulations and guidelines[258].
- Water Quality Standards: SH treatment technology must guarantee that the water released satisfies applicable water quality requirements established by regulatory bodies. This includes standards for SH limits, as well as other metrics like chemical oxygen demand, biochemical oxygen demand, and total suspended solids.
- Risk Assessment and Management: In order to determine the possible hazards connected to exposure to steroid hormones and to create effective risk management plans, such as source control procedures or extra treatment steps, regulatory bodies may demand risk assessments.
- Disposal of Residuals: It is a regulatory problem to properly dispose of treatment residuals, such as sludge or other byproducts containing concentrated SHs, in order to avoid contaminating the environment or people's exposure.

In addition to the above major highlights that are summarized in Fig. 16, there are no universal standards for the synthesis, characterization, or utilization of materials[259,257] used with these various techniques and the SHs removal techniques itself, leading to discrepancies in properties, procedures, and efficiency. This causes performance variations in SHs clean-up applications, which makes it challenging for practitioners and policymakers to adopt their use without reservation.

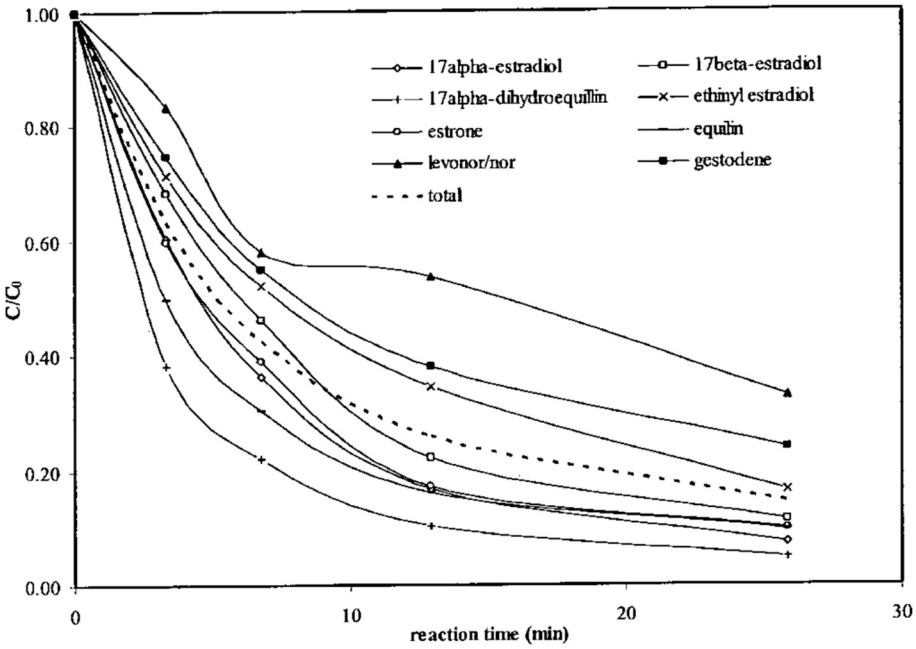

Fig. 15. Percentage degradation of SHs in clean water using sonolysis at 2 kW [254].

Fig. 16. Regulatory issues behind the use of various SH removal technologies.

8. Outlook and limitations

- More studies should be done to investigate the removal of steroids from water on a large scale. The efficiency of the systems should be determined also. Most published studies are research and pilot studies that are carried out in the laboratory under monitored conditions and factors.
- Most treatment technologies focus only on steroid hormones as a single contaminant, but natural wastewater is an aqueous mixture of organic, inorganic, and synthetic contaminants hence the need for more research in this area.
- Recent research methods and technologies focus on high percentage removal and degradation neglecting the cost-effectiveness, sustainability, energy efficiency, and toxicity of the intermediates resulting from the degradation.
- There is a dearth of research on techno-economic analysis and thus more work should be done on this as this is a focal point of interest for investors, engineers, industries, and policymakers.
- Studies should concentrate on boosting the effectiveness of the current conventional WWTPs steroid hormone removal by redesigning, altering operating factors, or upgrading them, as opposed to focusing primarily on advanced treatment methods. In addition, new methods of analysis need to be developed to detect synthetic hormones in WWTP. It will be useful for regulatory and standard organizations.
- There should be more studies on hybrid treatment technologies and their incorporation into WWTP for better performance.
- More work needs to be done on progesterone and androgen's origin and ecotoxicity because most research studies stress the environmental impact of estrogen.
- More research works reveal the ecotoxicity of steroid hormones even in very low concentrations, but more studies need to be done on its bioaccumulation and bioavailability.
- Due to its eco-toxicity even at low concentrations more constant and consistent environmental monitoring and research studies need to be done.

- There is a famine of work on the employment of bio-fabricated NPs for the photocatalytic degradation of SHs as only a few pieces of literature were found in this direction, and thus, more efforts should be devoted to that.

9. Conclusion

This paper overviewed steroid hormone (SH) compounds significant presence in the environment within a trace concentration, their characteristics source, health risk challenges, and advanced treatment strategies applicable, compared to conventional technologies that cannot offer a full treatment of the SH compounds. The advanced treatment technologies mentioned in this review comprise adsorption, membrane filtration, biochemical methods, biological approach, and AOP in tackling the scourge of non-biodegradable synthetic SH pollutants. All the treatment systems for SH removal, reviewed were applicable on a laboratory scale and can also be applied at the household level. From the review, the potential of biosorbent, and carbonaceous materials in the adsorptive removal of SH compounds also emerged, with a removal efficiency of up to 90% in a shorter time, which is superior to that of membrane filter system (~75%) and biological treatment that require a longer day.

Moreover, the membrane treatment approach was not viable for SH compound removal, due to their pore size being higher than SH compounds. However, the adoption of hybrid processes significantly ensures better SH compound removal than any of the single treatment procedures. The advanced oxidation processes comprising photocatalysis, Fenton process, electrocatalytic oxidation, and catalytic ozonation, evident higher removal efficiency of SH compound within a shorter time frame, and generation of mineralized products that are less toxic. This review improves our understanding of urgent SHs compound removal in WWTPs, aimed at mitigating provisions for freshwater, and identifies advanced treatment that requires improvement in SHs decontamination.

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Ajibola A. Bayode: Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Stephen Sunday Emmanuel:** Writing – review & editing, Writing – original draft. **Saheed O. Sanni**: Writing – original draft. **Olamide A. Olalekan:** Writing – original draft. **Odunayo Timothy Ore:** Writing – review & editing, Writing – original draft. **Daniel Terlanga Koko:** Writing – original draft. **Martins O. Omorogie**: Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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