G Model
HAZMAT-17653; No. of Pages 25
ARTICLE IN PRESS
Journal of Hazardous Materials xxx (2016) xxx–xxx
Contents lists available at ScienceDirect
Journal of Hazardous Materials
journal homepage: www.elsevier.com/locate/jhazmat

Progress in the biological and chemical treatment technologies for emerging contaminant removal from wastewater: A critical review

Mohammad Boshir Ahmed [a], John L. Zhou [a,*], Huu Hao Ngo [a], Wenshan Guo [a], Nikolaos S. Thomaidis [b], Jiang Xu [c]

[a] School of Civil and Environmental Engineering, University of Technology Sydney, Broadway, NSW 2007, Australia
[b] Department of Chemistry, University of Athens, Panepistimiopolis Zografou, 15771 Athens, Greece
[c] State Key Laboratory of Estuarine and Coastal Research, East China Normal University, 3663 North Zhongshan Road, Shanghai 200062, China

HIGHLIGHTS

- Emerging contaminant removal technologies have been summarized.
- MBR, microalgae and activated sludge are effective biological removal processes.
- Ozonation/H_2O_2 and photo-Fenton are highly effective chemical removal processes.
- Many hybrid systems have enhanced removal capacity of emerging contaminants.
- Future research regarding emerging contaminant removal has been proposed.

ARTICLE INFO

Article history:
Received 27 January 2016
Received in revised form 15 March 2016
Accepted 18 April 2016
Available online xxx

Keywords:
Emerging contaminants
Activated sludge
Photocatalysis
Ozonation
Hybrid systems

ABSTRACT

This review focuses on the removal of emerging contaminants (ECs) by biological, chemical and hybrid technologies in effluents from wastewater treatment plants (WWTPs). Results showed that endocrine disruption chemicals (EDCs) were better removed by membrane bioreactor (MBR), activated sludge and aeration processes among different biological processes. Surfactants, EDCs and personal care products (PCPs) can be well removed by activated sludge process. Pesticides and pharmaceuticals showed good removal efficiencies by biological activated carbon. Microalgae treatment processes can remove almost all types of ECs to some extent. Other biological processes were found less effective in ECs removal from wastewater. Chemical oxidation processes such as ozonation/H_2O_2, UV photolysis/H_2O_2 and photo-Fenton processes can successfully remove up to 100% of pesticides, beta blockers and pharmaceuticals, while EDCs can be better removed by ozonation and UV photocatalysis. Fenton process was found less effective in the removal of any types of ECs. A hybrid system based on ozonation followed by biological activated carbon was found highly efficient in the removal of pesticides, beta blockers and pharmaceuticals. A hybrid ozonation-ultrasound system can remove up to 100% of many pharmaceuticals. Future research directions to enhance the removal of ECs have been elaborated.

1. Introduction

Emerging contaminants (ECs) are primarily synthetic organic chemicals that have been recently detected in natural environments [1–3]. ECs are a large and relatively new group of unregulated compounds [4] and can potentially cause deleterious effects in aquatic and human life at environmentally relevant concentrations which are becoming a growing concern [1,5,6]. They are the ingredients mostly detected in municipal sewage, daily household products, pharmaceutical production plants, wastewater, hospitals, landfills, and natural aquatic environment [7–9]. ECs concentration may range from a few ng L^{-1} to a few hundred μg L^{-1} [8,10]. Such concentrations in the aquatic environment may cause ecological risk such as interference with endocrine system of high organisms, microbiological resistance, and accumulation in soil, plants and animals [11], as these ECs are not completely removed by conventional wastewater treatment processes [6,12,13]. ECs include mostly pharmaceutical organic contaminants, personal care products (PCPs), endocrine disrupting compounds (EDCs),

* Corresponding author.
E-mail address: junliang.zhou@uts.edu.au (J.L. Zhou).

http://dx.doi.org/10.1016/j.jhazmat.2016.04.045
0304-3894/

surfactants, pesticides, flame retardants, and industrial additives among others.

Pharmaceutical organic contaminants and PCPs include analgesics, lipid regulators, antibiotics, diuretics, non-steroid anti-inflammatory drugs (NSAIDs), stimulant drugs, antiseptics, analgesic, beta blockers, antimicrobials, cosmetics, sun screen agents, food supplements, fragrances and their metabolites and transformation products. They can affect water quality and potentially affect drinking water supplies, ecosystem and human health [13–15]. Their environmental bioaccumulation exacerbates the abnormal hormonal control causing reproductive impairments, decreased fecundity, increased incidence of breast and testosterone cancers, and persistent antibiotic resistance [16]. Of particular concern are antibiotic residues which can induce the development of antibiotic resistant genes potentially favouring superbugs [6].

EDCs are exogenous substances or mixtures that alter the functions of the endocrine systems and consequently cause adverse health effect in an intact organism, or its progeny or populations [17]. The effects associated with EDCs are breakage of eggs of birds, fishes and turtles, problems in reproductive systems, change in immunologic system of marine mammals, reduction of sperm of human organ, increase in the incidence of breast, testicle and prostate cancers, and endometriosis [14]. Pesticides have immune-depressive effects in fishes, mammals and can modify haemopoietic tissue of anterior kidney [18]. Surfactants can affect physical stability of human growth hormone formulations and are responsible for the endocrine activity [19].

The potential long-term effects of ECs in water are still uncertain and need further investigation. At present, different government and non-government organizations including the European Union (EU), the North American Environmental Protection Agency (EPA), the World Health Organization (WHO), or the International Program of Chemical Safety (IPCS) are considering these problems and setting up directives and legal frameworks to protect and improve the quality of freshwater resources [14].

A variety of different physical, chemical and biological technologies have already been used to remove or degrade the residues of ECs over the last few decades [20,21]. Biological treatment technologies are by far the most widely used for ECs removal, including activated sludge, constructed wetland, membrane bioreactor (MBR), aerobic bioreactor, anaerobic bioreactor, microalgae bioreactor, fungal bioreactor, trickling filter, rotating biological reactor, nitrification, enzyme treatment and biosorption. It has been reported that some non-biodegradable organic micropollutants cannot be sufficiently removed using biological treatment processes. Chemical treatment technologies are also widely used for the degradation of these micropollutants, including conventional oxidation methods such as Fenton, ozonation, photolysis and advanced oxidation processes (AOPs) such as ferrate, photo-Fenton, photocatalysis, solar driven processes, ultra sound process, and electro-Fenton process. Moreover, some hybrid systems have recently been applied to enhance the removal of a wide range of ECs. The advantages and challenges of different processes for the removal of ECs are outlined in Table 1.

The majority of polar and semi polar pesticides and pharmaceuticals will remain partitioned in the aqueous phase due to their relatively high water solubility, hence their removal by physical processes such as sedimentation and flocculation is not effective [22], and has been reported to be less than 10% [23,24]. Thus further discussions of those processes are not reviewed here. The discussion of other physical treatment processes such as membrane, reverse osmosis, ultrafiltration, microfiltration, nanofiltration and adsorption processes is also excluded from this review, although these physical processes can be part of hybrid or integrated treatment technologies for ECs removal.

Thus, the aim of this review is to critically evaluate the viability of biological, chemical, and hybrid treatment processes as a means to remove ECs from wastewater. Specifically the article provided a summary of effectiveness of different wastewater treatment processes for ECs removal, discussed conventional wastewater treatment processes along with advance and hybrid treatment processes for ECs removal, and discussed the challenges and the current knowledge gaps limiting the effectiveness of biological and chemical treatment processes. Some of the future research directions have also been suggested.

2. Biological treatment technologies

Biological treatment technologies have been widely applied for the removal of ECs predominantly by the mechanism of biodegradation. Biodegradation is the process by which large molecular weight ECs are degraded by microorganisms such as bacteria, algal and fungi into small molecules [4], and even biomineralised to simple inorganic molecules such as water and carbon dioxide. In conventional biodegradation process, microorganisms use organic compounds as primary substrates for their cell growth and induce enzymes for their assimilation [10]. Some ECs are toxic and resistant to microbial growth hence inhibiting biodegradation, in which case a growth substrate is needed to maintain microbial growth for biodegradation, a process known as cometabolism [10]. Biodegradation methods have traditionally been used in wastewater treatment systems for the removal of ECs. They can be divided into aerobic and anaerobic processes. Aerobic applications include activated sludge, membrane bioreactor, and sequence batch reactor. Anaerobic methods include anaerobic sludge reactors, and anaerobic film reactors. The wastewater characteristics play a key role in the selection of biological treatments [7,28]. The wastewater treatment processes can be broadly classified as conventional processes and non-conventional processes, which are described in subsequent sections.

2.1. Progress and challenges in conventional treatment processes

Removal or degradation capacity of ECs depends on the chemical and biological persistence of ECs, their physicochemical properties, the technology used, and operation conditions. For the highly polar substances e.g. most pharmaceuticals and their corresponding metabolites, the most important removal process is through the biological transformation or mineralization by microorganisms. The removal rates strongly depend upon the treatment technology, the operation conditions, and target contaminants [36]. The identification of degradation products in environmental samples is a challenging task because not only are they present at very low concentrations but also they are present in complex matrices that may interfere with detection [36,37].

2.1.1. Biological trickling filter and biofilm reactor

A biological trickling filter is a three-phase system with fixed biofilm carriers. Wastewater enters the bioreactor through a distribution zone, trickles downward over the biofilm surface, and air moves upward or downward in the third phase [38]. Bio-trickling filters have been used in wastewater treatment plants (WWTPs) for decades in the removal of biochemical oxygen demand (BOD), chemical oxygen demand (COD), pathogen decontamination, odor and air pollution control, but their application to ECs removal has not become wide practice [39–41]. Trickling filters or biobeds were used either alone or in combination with other treatment processes such as activated sludge. Some bio-processes such as activated sludge, aerated lagoon and trickling filters have reported very different removal efficiencies from almost complete removal