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Review

Application of ozonation for pharmaceuticals and personal care products removal from water

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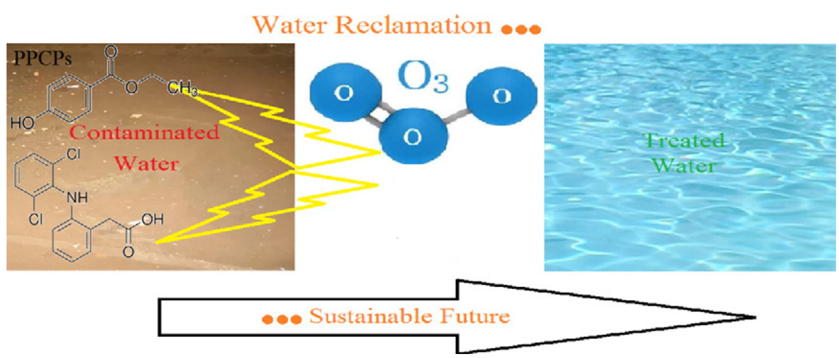
HIGHLIGHTS

- Reclaimed wastewater is becoming an important source of fresh water.
- Pharmaceuticals and personal care products are a concern on quality of reused water.
- Ozone removes such compounds even if with low mineralization.
- Ozonation is a suitable alternative specially when integrated with biofiltration.
- Ozonation is compared with other alternatives for water recovery.

GRAPHICAL ABSTRACT



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ABSTRACT

Due to the shortening on natural water resources, reclaimed wastewater will be an important water supply source. However, suitable technologies must be available to guaranty its proper detoxification with special concern for the emerging pharmaceutical and personal care products that are continuously reaching municipal wastewater treatment plants. While conventional biological systems are not suitable to remove these compounds, ozone, due to its interesting features involving molecular ozone oxidation and the possibility of generating unselective hydroxyl radicals, has a wider range of action on micropollutants removal and water disinfection. This paper aims to review the studies dealing with ozone based processes for water reuse by considering municipal wastewater reclamation as well as natural and drinking water treatment. A comparison with alternative technologies is given. The main drawback of ozonation is related with the low mineralization achieved that may lead to the production of reaction intermediates with toxic features. The use of hydrogen peroxide and light aided systems enhance ozone action over pollutants. Moreover, scientific community is focused on the development of solid catalysts able to improve the mineralization level achieved by ozone. Special interest is now being given to solar light catalytic ozonation systems with interesting results both for chemical and biological contaminants abatement. Nowadays the integration between ozonation and sand biofiltration seems to be the most interesting cost effective methodology for water treatment. However, further studies must be performed to optimize this system by understanding the biofiltration mechanisms.

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Contents

1. Introduction

Modern society requirements are leading to an increasingly stress over the natural resources. Projections reveal that by 2050, water, energy and food demands will increase by 55%, 80% and 70% respectively compared with current values (Loeb, 2016). Moreover, the fresh water bodies are continuously affected by contamination from municipal and industrial polluted streams. This is leading to a great threat to ecosystems and human health (Ganiyu et al., 2015).

The shortening on the amount of water supply sources and their quality is pushing towards the spreading of the implementation of municipal wastewater (MWW) reclamation and reuse strategies (Bixio et al., 2006). In this ambit, in the last decade attention has been driven to the occurrence of persistent organic compounds in municipal wastewaters, natural waters and even drinking water, such as pharmaceuticals and personal care products - PPCPs (Rivera-Utrilla et al., 2013; Noguera-Oviedo and Aga, 2016). Among these can be, for example, referred the organic UV filters (Molins-Delgado et al., 2016a), pharmaceutical active compounds, endocrine disrupting compounds and artificial sweeteners (McKie et al., 2016). These contaminants are found in secondary MWW in the range of $ng \cdot L^{-1}$ to $\mu g \cdot L^{-1}$ (Petrovic et al., 2005; Hollender et al., 2009; Ahmed et al., 2017). The presence of such compounds is related with the incorrect disposal of unused drugs and personal care products in the sewage but also due to human excretion (Heberer, 2002). Some specific compounds such as pesticides, dioxins and dioxins-like, aromatic hydrocarbons, flame retardants and industrial compounds were classified by European Union as priority substances (Directive, 2013). Besides, a group of 10 substances are recommended to be monitored (Decision, 2015).

The traditional methodologies applied in municipal wastewater treatment plants (MWWTP) are unable to remove emerging contaminants (Martínez-Bueno et al., 2007). Even if some compounds (e.g. ibuprofen, paracetamol, estradiol, acetyl salicylic acid) are known to be biodegradable, others such as diclofenac, parabens, carbamazepine, cromiton, diazepam are not removed biologically and their concentration is maintained after treatment (Joss et al., 2008; Onesios et al., 2009; Abdelmelek et al., 2011). Some others such as bisphenol-A, bezafibrate and naproxen can be removed by activated sludge after a high retention time (Clara et al., 2005). The removal of pharmaceutical and personal care products (PPCPs) in the municipal wastewater treatment plants depends largely on the compounds hydrophobicity (Rosal et al., 2010a) as well as on their dissociation constant. For example, Espejo et al. (2014) verified during biological treatment of primary municipal wastewater spiked by nine pharmaceuticals that for a hydraulic retention time of 24 h only acetaminophen and caffeine were totally removed. Metoprolol, sulfamethoxazole and hydrochlorothiazide were partially removed while antipyrine, carbamazepine, diclofenac and ketorolac were totally biorefractory.

The continuous discharge from MWWTP into the environment is a source of persistent compounds. Thus they are reaching and accumulating in the natural water courses/rivers which is an increasing concern for these streams that are used as water sources for human applications as, for example, drinking water source through indirect potable reuse (Lee and von Gunten, 2010). Moreover, MWW treated water is starting to be considered an important component of water resources supply (Li et al., 2015). So far, no direct relation was found between the low concentrations of these chemicals found in these streams and human health effects. However, the concern related with their potential chronic health effects due to long term exposure is increasing as no reliable data are still available about this emerging environmental danger (Stackelberg et al., 2004). Thus, the precautionary principle imposes that those compounds must be removed from human use water (Huber et al., 2003). It must be especially given attention to the possible synergetic or additive effect of mixtures of pollutants (Schwarzenbach et al., 2006).

Some of these compounds are considered as endocrine disrupting chemicals (EDCs) which are substances able to interfere with the endocrine system. The exposure to such substances is a matter of public concern since they are alleged to be related with some tumours (Molins-Delgado et al., 2016b) as well as reproductive problems (Esplugas et al., 2007). On the other hand the spread of antibiotics throughout the ecosystems leads to the proliferation of antibiotic-resistant bacteria which easily disseminate posing serious human health danger (Chen et al., 2016). Antibiotics are considered as pseudo-persistent in the environment due to their continuous discharge (Watkinson et al., 2009). It should also be stressed that, although MWWTP are able to remove up to 2 logarithmic cycles of bacteria, do not prevent antibiotic resistance prevalence and it is estimated that lead to the disposal of > 109 antibiotic resistant coliforms per minute to the environment (Michael et al., 2013; Vaz-Moreira et al., 2014) which also constitutes an important health issue.

World Health Organization (WHO) and International Water Association (IWA) recommend that a preventive risk management must be