Solar photo-Fenton and adsorption on activated carbon for the removal of antibiotics, antibiotic resistance determinants and toxicity from urban wastewater

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The presence of contaminants of emerging concern such as antibiotics, antibiotic-resistant bacteria (ARB) and resistance genes (ARG) in wastewater effluents and the inability of the conventional wastewater treatment processes (i.e. conventional activated sludge) to completely eliminate them is one of the most challenging aspects of wastewater reuse. Advanced Oxidation Processes (AOPs), based on the *in situ* generation of hydroxyl radicals ($HO^{\bullet}$) with high efficiency in degrading various microcontaminants, have gained considerable attention the last years with regard to their real-scale application in urban wastewater treatment plants. Among the various AOPs, photo-Fenton is an attractive option for the treatment of wastewater effluents due to its environmentally friendly application and the prospect of operating under natural solar irradiation (i.e. solar photo-Fenton), hence lowering the operating cost of the process considerably (Badawy *et al*., 2009). Activated Carbon (AC) adsorption can enhance further the removal of the microcontaminants, as well as reduce the toxicity of the AOPs-treated wastewater due to the potential adsorption of toxic oxidation products. Within this context, this work aimed at exploring the efficiency of the solar photo-Fenton process at a pilot scale, in removing selected antibiotics and ARB, as well as toxicity from secondary treated wastewater effluents. The degradation of a mixture of antibiotics, namely ampicillin (AMP), clarithromycin (CLA), erythromycin (ERY), ofloxacin (OFL), tetracycline (TET), trimethoprim (TMP) and sulfamethoxazole (SMX), was investigated by testing various oxidant (H_2O_2) concentrations. Ferrous iron concentration was kept constant in all experiments (5 mg/L), to avoid the requirement for iron removal at the end of the process according to the iron discharge limits set in the existing regulation (Cyprus Law, 106(I)/2002). The phyto- and eco-toxicity of the treated samples were also evaluated against three plant species (*Sorghum saccharatum*, *Lepidium sativum*, *Sinapis alba*) and a crustacean (*Daphnia magna*), respectively. Moreover, the disinfection potential of the solar photo-Fenton process as to the inactivation of frequently encountered bacteria in wastewater, which have the potential to harbour antibiotic resistance to selected antibiotics, i.e. *Escherichia coli*, *Pseudomonas aeruginosa*, *Enterococcus* and total heterotrophic bacteria was assessed. Finally, the capacity of solar photo-Fenton was assessed with regard to the inactivation of selected ARG (e.g 16S rRNA, *bla*TEM, *bla*OXA, *bla*CTX, *tet*M, *sul*1, etc).The obtained results demonstrated the ability of the solar photo-Fenton process to completely degrade all the tested antibiotics under the optimum experimental conditions ([Antibiotics]=100 μg/L; $[Fe^{2+}]$=5 mg/L; $[H_2O_2]$=100 mg/L; pH=2.8-2.9, $t_{30W,n}$=115 min). The photocatalytic removal of all antibiotics followed pseudo-first-order kinetics, with AMP being degraded faster than the other antibiotics. Interestingly, all colonies harbouring resistance to OFL, TMP and ERY apparently survived solar photo-Fenton process, while prolonged treatment time was needed for their complete elimination. Phyto- and eco-toxicity tests showed increased toxicity until 120 min of treatment, induced either by the oxidation of the the dissolved effluent organic matter originally present in wastewater, or the oxidation of the parent antibiotics to more toxic transformation products. Since the ecotoxicity of the treated samples at the end of the oxidation process was similar with that of the untreated wastewater, additional adsorption experiments using granular activated carbon (GAC) were carried out to investigate whether the residual toxicity could be further reduced. The results revelaed that the post-treatment of the solar photo-Fenton treated flow with 500 mg/L of GAC resulted in complete elimination of ecotoxicity within 15 min of contact time, indicating that the toxic oxidation products were rapidly adsorbed onto the GAC surface. Finally, the findings of this study demontrated the capacity of the solar photo-Fenton to inactivate the selected ARG.

References

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