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Research article

Paracetamol and salicylic acid removal from contaminated water by microalgae☆

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ABSTRACT

The biomass growth, pharmaceutical removal and light conversion efficiency of *Chlorella sorokiniana* under the presence of paracetamol (PC) and salicylic acid (SaC) were assessed and compared at two different concentrations of these pharmaceuticals (I: 25 mg l^{-1}, II: 250 mg l^{-1}). Microalgae were resistant to these concentrations and, moreover, their growth was significantly stimulated ($p \leq 0.05$) under these drugs (biomass concentration increased above 33% PCI, 35% SaCI, 13% PCII and 45% SaCII, as compared with the respective positive controls). At the steady state of the semicontinuous culture, *C. sorokiniana* showed removal efficiencies above 41% and 69% for PCI and PCII, respectively; and above 93% and 98% for SaCI and SaCII, respectively. Under an irradiance of 370 μE m^{-2} s^{-1}, higher quantum yields were reached by microalgae under the presence of drugs, either at dose I or II, than by the respective positive controls. These results point to *C. sorokiniana* as a robust strain for the bioremediation of paracetamol and salicylic acid concentrated wastewaters.

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1. Introduction

The global increase of the production and use of drugs has led to a growing international awareness on the disposal of pharmaceutical wastes due to their potential deleterious effects (Halling-Sorensen et al., 1998). In the aquatic environment, the presence of pharmaceuticals may cause physiological responses in organisms for which they were not intended, such as accumulation in tissues, reproductive damage, inhibition of cell proliferation and behavioural changes (Escher et al., 2011).

Pharmaceuticals are known to reach the environment through usage and/or inappropriate disposal (Khetan and Collins, 2007). Therefore, main sources of pharmaceuticals in the environment include hospitals (Gupta et al., 2009; Suárez et al., 2009; Escher et al., 2011; Verlicchi et al., 2012), landfill leachates (Andrews et al., 2012; Clarke et al., 2015) and effluents from sewage treatment plants (STPs), which is probably the most investigated route of entry. Still, some authors (Larsson et al., 2007; Carlsson et al., 2009; Larsson and Fick, 2009) have pointed to the highly concentrated effluents from drugs production facilities as important sources of pharmaceuticals to the environment. These concentrated effluents should be appropriately treated to remove active pharmaceuticals ingredients (APIs) before discharge, either in natural waters or in the municipal sewage system.

To date, the nonexistence of limiting regulations (Bolong et al., 2009; Barceló and Petrovic, 2006) has led to a lack of control on the discharge of APIs by manufacturing facilities (Larsson and Fick, 2009). However, given public concern about the presence of pharmaceuticals in the environment, it is expectable that regulations will come out in the near future. Yet, conventional wastewater treatments were designed to eliminate carbon, nitrogen and phosphorus, but not to remove APIs, which pose high biological activity at low concentrations, are mostly hydrophilic and have low adsorption rates (Martz, 2012).

Advanced oxidation processes (AOPs) such as oxidation, ozonation, perozonation, direct photolysis, TiO_2 photocatalysis, solar photocatalysis, Fenton reactions and ultrasonic irradiation have been pointed as promising treatments for the removal of APIs from wastewater (Deegan et al., 2011). Nevertheless, the high operational costs, the harsh reaction conditions and the generation of

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secondary pollutants are important disadvantages of this sort of treatments (Wu et al., 2012). Conversely, biodegradation treatments are considered as a low-cost and environmentally friendly option (Hasan et al., 2011; Chen et al., 2010). Although there are preliminary studies about biodegradation of pharmaceuticals by microorganisms (Combarros et al., 2014; Zhang et al., 2013; Wu et al., 2012; De Gusseme et al., 2011), very few studies have been carried out on the removal of pharmaceuticals by microalgae (Gupta et al., 2015 and references within). On the contrary, microalgae capacity to remove nutrients from wastewater is well known, Choudhary et al. (2016) having highlighted the importance of the selection of robust strains/consortia that can efficiently grow under concentrated wastewaters. However, to our best knowledge, there are not published results on microalgae resistance and efficiency under extreme concentrations of drugs, which may be expected in wastewaters from pharmaceutical industry.

In this context, and after having proved the capability of *Chlorella sorokiniana* to remove pharmaceuticals from water (Escapa et al., 2015), the aim of this work was to assess microalgae response towards relatively high drugs concentration. For this purpose, microalgae growth kinetics, pharmaceutical removal rates and light conversion efficiency under paracetamol and salicylic acid were evaluated and compared at two different concentrations of these drugs, which are among the most consumed and found in natural waters worldwide. In the aquatic environment, paracetamol is easily accumulated due to its relative high solubility and hydrophilicity (Muir et al., 1997). Regarding salicylic acid, its toxicity and tendency to accumulate in the environment are well known (Combarros et al., 2014). Therefore, results obtained in this work are expected to contribute to the knowledge on the application of microalgae on the bioremediation of wastewater, namely on the removal of emerging contaminants such as pharmaceuticals.

2. Materials and methods

2.1. Microorganism and culture conditions

The microalgae strain used in this study was *Chlorella sorokiniana CCAP 211/8 K (UTEX Culture Collection)*. Inoculum for the experiments was cultivated in the standard culture medium Mann and Myers (Mann and Myers, 1968) in 250 ml Erlenmeyer flasks. The culture medium is composed of (grams per litre of distilled water): 1.2 $MgSO_4 \cdot 7H_2O$, 1.0 $NaNO_3$, 0.3 $CaCl_2$, 0.1 K_2HPO_4, 3.0×10^{-2} Na_2EDTA, 6.0×10^{-3} H_3BO_3, 2.0×10^{-3} $FeSO_4 \cdot 7H_2O$, 1.4×10^{-3} $MnCl_2$, 3.3×10^{-4} $ZnSO_4 \cdot 7H_2O$, 7.0×10^{-6} $Co(NO_3)_2 \cdot 6H_2O$, 2.0×10^{-6} $CuSO_4 \cdot 5H_2O$. The *C. sorokininana* inoculum was maintained inside a vegetal culture chamber, where the growth conditions remained constant, under controlled temperature (25 ± 1 °C), irradiance (175 μE m^{-2} s^{-1}), photoperiod (12:12) and stirring (250 rpm).

Experiments were carried out in bubbling column photobioreactors (PBRs) with spherical bases (40 mm diameter and 300 mm height with 300 ml capacity), keeping an operating volume of 250 ml. PBRs containing Mann and Myers medium were inoculated with the above inoculum of *C. sorokiniana* in order to have an initial microalgae concentration of about 3.2×10^6 cells ml^{-1}. Then, culture conditions were kept constant under controlled temperature (25 ± 1 °C), irradiance (370 μE m^{-2} s^{-1}) and photoperiod (12:12), inside a vegetal culture chamber. PBRs were illuminated by 8 fluorescent lamps (58 W, 2150 lumen, Philips, France) and aerated at a rate of 0.3 v/v/min with CO_2 enriched at 7% v/v, which was injected on demand to keep a constant pH (pH = 7.5 ± 0.5) as controlled by a pH sensor. Before injection, the air was filtered through 0.2 μm sterile air-venting filter (Millex-FG50, Millipore). At the beginning of the experiments, the culture medium, the PBRs and the tubing for the supply of air were sterilized in autoclave (121 °C, 1 atm, and 20 min) to avoid contaminations. In the experiments under the presence of paracetamol (PC) or salicylic acid (SaC), these drugs were added to the sterilized culture medium but not submitted to the sterilization process so to guarantee their stability.

2.2. Experimental set-up

PBRs were operated in batch mode until the end of the exponential growth phase. Then, the operation mode was semicontinuous with a daily renewal rate of 30%. Therefore, under semicontinuous operation, 30% of the reactor culture volume was daily taken off and replaced by fresh medium (containing 25 mg l^{-1} of either paracetamol or salicylic acid, in the case of PC and SaC treatments, respectively). PBRs were operated in semicontinuous mode until stability of the growth parameters at the steady state.

The experiments carried out consisted of inoculated culture medium together with paracetamol (PC) or salicylic acid (SaC). Two different and non-simultaneous sets of experiments were run under two different concentrations of these pharmaceuticals: (i) dosage I (25 mg l^{-1}), as described by Escapa et al. (Escapa et al., 2015), which were named as PCI and SaCI; and (ii) dosage II (250 mg l^{-1}), named as PCII and SaCII, respectively. For each set of experiments, positive controls with inoculated culture medium without drugs (C+I and C+II, respectively) were run in parallel. Also, negative controls with not inoculated culture medium but PC or SaC were run simultaneously with each concentration set of experiments (C_P-I, C_{Sa}-I and C_P-II, C_{Sa}-II, respectively). The treatments and the abbreviations used in this work are displayed in Table s1 (as supplementary material). Three simultaneous replicates of each treatment were operated under identical conditions for each set of experiments. Paracetamol ($C_8H_9NO_2$, ≥99%) was supplied by Roic Pharma and salicylic acid ($C_7H_6O_3$, ≥99%) by Panreac. Physico-chemical properties of these pharmaceuticals are displayed in Table s2 (as supplementary material).

Throughout the experiments, the culture growth was daily monitored by biomass concentration and cell density. Also, to assess the pharmaceuticals removal, the concentration of paracetamol and salicylic acid was daily monitored. In addition, the irradiance in the absence of cells in the central point inside of the PBR was measured to evaluate the light conversion efficiency of the cells.

2.3. Analytical methods

Biomass concentration (C_b) was determined by optical density at 680 nm (OD_{680}) by spectrophotometric (UV/visible spectrophotometer BECKMAN DU640) and verified by dry weight. Preliminary studies were conducted to determine OD_{680} as a function of dry weight as shown in Eq. (1):

$$OD_{680} = 5.1834 \times C_b + 0.0128, \quad R^2 = 0.9983 \tag{1}$$

For the determination of dry weight, 10 ml of culture were filtered through a 0.45 μm Whatman filter, which was then washed with 20 ml HCl (0.5 M) to dissolve precipitated salts, and dried in an oven at 80 °C for 24 h. Dry weight was then obtained by mass difference of the filter after and before filtration. Additionally, the growth of the culture was measured as cell density (*Nc*) by cell counting with a Neubauer chamber.

A Waters HPLC 600 equipped with a 2487 dual λ absorbance detector was used for the quantification of the target pharmaceuticals. The wavelengths of detection were 246 and 236 nm for paracetamol and salicylic acid, respectively. A Phenomenex C18

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