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Removal of pharmaceuticals from wastewater by fungal treatment and reduction of hazard quotients

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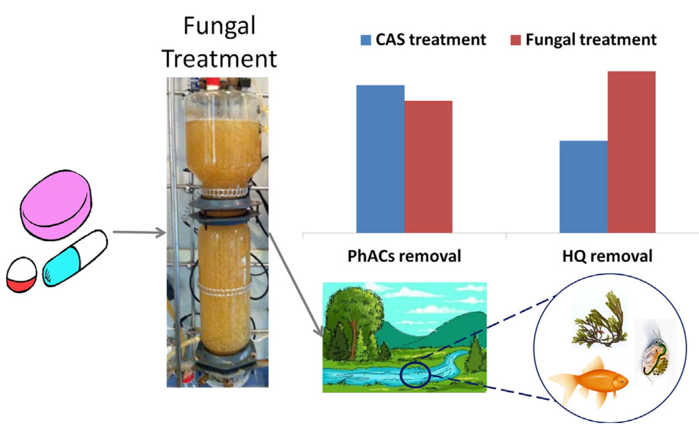
HIGHLIGHTS

- Efficiency of fungal treatment was evaluated in different wastewater effluents.
- Removal of up to 81 pharmaceuticals in all treatments was estimated.
- Reduction of environmental risk after fungal treatment was also assessed.
- Antibiotics and psychiatric drugs were better removed by fungal treatment.
- Fungal treatment showed promising results from an ecotoxicological point of view.

GRAPHICAL ABSTRACT



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ABSTRACT

The elimination of 81 pharmaceuticals (PhACs) by means of a biological treatment based on the fungus *Trametes versicolor* was evaluated in this work. PhAC removal studied in different types of wastewaters (urban, reverse osmosis concentrate, hospital, and veterinary hospital wastewaters) were reviewed and compared with conventional activated sludge (CAS) treatment. In addition, hazard indexes were calculated based on the exposure levels and ecotoxicity for each compound and used for the evaluation of the contaminants removal. PhAC elimination achieved with the fungal treatment (mean value 76%) was similar or slightly worse than the elimination achieved in the CAS treatment (85%). However, the fungal reactor was superior in removing more hazardous compounds (antibiotics and psychiatric drugs) than the conventional activated sludge in terms of environmental risk reduction (93% and 53% of reduction respectively). Fungal treatment can thus be considered as a good alternative to conventional treatment technologies for the elimination of PhACs from wastewaters.

1. Introduction

Over the last years a wide range of pharmaceutically active compound (PhAC) residues have been found in several environmental matrices (Gibs et al., 2007; Heath et al., 2010; Loos et al., 2010; da

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Silva et al., 2011; Yang et al., 2011) due to their extensive consumption and pseudo-persistence in the environment (Joss et al., 2005; Petrović et al., 2005; Han et al., 2006; Gros et al., 2010; Verlicchi et al., 2012b). Several studies suggested that conventional activated sludge (CAS) technologies used in urban wastewater treatment plants (WWTPs) are not effective enough to eliminate PhACs, since they are not designed to remove complex compounds (Verlicchi et al., 2012b; Frédéric and Yves, 2014). As a consequence, some innovative wastewater treatment technologies have been developed in order to achieve higher removal efficiency of this type of pollutants (Escolà Casas et al., 2015a, 2015b; Bagheri et al., 2016; Ferre-Aracil et al., 2016; Ng et al., 2016). Among them, the fungal treatment of wastewaters has been highlighted as a promising technology because of the unspecific enzymatic system of lignolytic fungi, which is able to degrade a wide range of PhACs, even though they are present at very low concentrations (Marco-Urrea et al., 2009; Rodríguez-Rodríguez et al., 2012; Gros et al., 2014; Llorens-Blanch et al., 2015).

Besides monitoring the reduction of PhAC concentration, a useful tool to evaluate the efficiency of the fungal treatment is measuring the environmental impact on aquatic organisms of the decrease in PhAC concentration. This could be achieved by environmental risk assessment (ERA), which estimates the probability of a compound to cause undesired environmental effects (Carlsson et al., 2006) based on both, concentration and ecotoxicity of each particular compound. Many studies have assessed the environmental risk of the PhACs in several treated wastewater effluents (Gros et al., 2010; Escher et al., 2011; Al Aukidy et al., 2012; Santos et al., 2013; Collado et al., 2014; Kosma et al., 2014), but only one has considered the efficiency of CAS treatment in ecotoxicological terms (Verlicchi et al., 2012a); and none has used this approach to evaluate alternative wastewater technologies, such as those based on fungal treatment. In this work we present the use of ERA as a complementary tool to evaluate effectiveness of fungal treatment in comparison to a CAS treatment. Four different types of wastewaters treated with the fungal treatment were considered (Cruz-Morató et al., 2013; Badia-Fabregat, 2014; Cruz-Morató et al., 2014; Badia-Fabregat et al., 2015a, 2015b); and treatment performance was evaluated using not only the 81 PhAC removal efficiency but also the environmental risk associated. Results were compared to those obtained from CAS treatment of urban wastewater (Collado et al., 2014), used as reference treatment.

2. Materials and methods

2.1. Water samples

Wastewaters considered in this study were i) wastewater effluents from a university village, considered as urban wastewater, ii) reverse osmosis concentrate (R.O. concentrate) from urban WWTP effluent, with a 34% of rejection rate (Badia-Fabregat, 2014; Badia-Fabregat et al., 2015a), iii) hospital wastewater and iv) veterinary hospital wastewater; whereas v) conventional urban wastewater was obtained from a municipal WWTP from a town located in North-east Spain (20,000 equivalent inhabitants, 2.100 m^3 d^{-1} volume treated; with a hydraulic retention time (HRT) of 48 h and a sludge retention time (SRT) of 20–22 days; Collado et al., 2014). Details about wastewater types and treatments specifications are reported in Table 1. Data from the monitoring study by Collado et al., 2014 was used as reference values of CAS treatment since both, their study and our fungal treatment experiments, targeted the same set of PhACs (see Table S1) using the same analytical methodology (Gros et al., 2012). Levels of these compounds along the treatment in the WWTP were measured in three different seasons of the year in dry weather conditions: in May 2011 (16th–20th), January 2012 (16th–20th) and August 2012 (6th–10th). Every seasonal campaign 48-h composite and flow proportional samples were collected both, at the WWTP inlet (before the primary treatments) and at the outlet of the secondary treatment, by means of an auto-sampler. Each sample was analyzed in triplicate. PhAC concentrations and removal values obtained in this study were consistent with the values found in the literature for other urban WWTPs (Verlicchi et al., 2012b) and thus considered representative of conventional WWTP. Different operational parameters for the fungal treatment were tested (batch and continuous operation, nutrients addition, treatment time, etc.; Table 1) in order to maximize the degradative capacity of PhACs. An overview of the PhAC concentrations before and after the corresponding treatment can be found in Table S2. Removal data achieved for PhACs with the fungal treatment under the different treatment conditions and with the different type of effluents considered was compared with that obtained with CAS treatment. Even though the fungal treatments were performed at lab-scale and the operational parameters varied from one treatment to the other, a comparison with a full-scale CAS can provide a preliminary idea about the efficiency and potential of the fungal treatment.

2.2. Fungal reactor

Trametes versicolor (ATCC#42530) was obtained from the American Type Culture Collection. Maintenance and pellet production were done as previously described by Blanquez et al. (2004). Pellets of *T. versicolor* were added at approximately 2 g dry cell weight (DCW) L^{-1}. Temperature was set up at 25 °C and pH was controlled to be constant at 4.5 ± 0.5 by adding HCl 1 M or NaOH 1 M. Glucose was supplied, together with ammonia tartrate, in pulses of 0.6 min h^{-1} from a concentrated stock solution at a final rate of 552 and 1.24 mg g^{-1} d^{-1} respectively; the addition of a little amount of nutrients has been proved to enhance the efficiency of the fungal treatment (Badia-Fabregat et al., 2015a). Samples for the analysis of PhACs were obtained by a silicone tube and were kept in sterilized glass vials. They were vacuum filtered with 1.2 μm Wathman GF/C filters followed by 0.45 μm nylon filter (Millipore). Sample concentration and dilution caused by acid/base addition were taken into account for PhAC removal calculations. Details about specific operational parameter can be found in the articles referred in Table 1.

Table 1
Wastewater samples considered in the present study.

Samples	Treatment	Reactor type	Sterile influent	Nutrients input	Treatment time	Reference
Urban wastewater	CAS	Continuous	No	No	2 days	Collado et al. (2014)
University village wastewater I	Fungal treatment	Batch	Yes	Yes	8 days	Cruz-Morató et al. (2013)
University village wastewater II	Fungal treatment	Batch	No	Yes	8 days	Cruz-Morató et al. (2013)
Reverse osmosis concentrate I	Fungal treatment	Batch	Yes	Yes	6 days	Badia-Fabregat et al. (2015a)
Reverse osmosis concentrate II	Fungal treatment	Continuous	No	Yes	6 days	Badia-Fabregat (2014)
Hospital wastewater I	Fungal treatment	Batch	Yes	Yes	8 days	Cruz-Morató et al. (2014)
Hospital wastewater II	Fungal treatment	Batch	No	Yes	8 days	Cruz-Morató et al. (2014)
Veterinary hospital I	Fungal treatment	Batch	No	Yes	14 days	Badia-Fabregat et al. (2015b)
Veterinary hospital II	Fungal treatment	Continuous	No	Yes	8 days	Badia-Fabregat et al. (2015b)