International Biodeterioration & Biodegradation 100 (2015) 79–84

Contents lists available at ScienceDirect

International Biodeterioration & Biodegradation

journal homepage: www.elsevier.com/locate/ibiod

Short communication

Elimination and detoxification of fungicide miconazole and antidepressant sertraline by manganese peroxidase-dependent lipid peroxidation system

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ARTICLE INFO

Article history:
Received 28 August 2014
Received in revised form
4 February 2015
Accepted 11 February 2015
Available online 5 March 2015

Keywords:
Miconazole
Sertraline
Elimination
Manganese peroxidase
Unsaturated fatty acids
Lipid peroxidation

ABSTRACT

The imidazole fungicide miconazole (MCZ) and the antidepressant sertraline (SER) have the potential to adversely affect aquatic organisms when they are released into the environment. To assess if they could be broken down, MCZ and SER were treated with laccase and manganese peroxidase (MnP) from lignin-degrading white-rot fungi. The MnP-dependent lipid peroxidation system with Tween 80 containing unsaturated fatty acid was effective in eliminating MCZ and SER; the MnP-Tween 80 system helped eliminate 88% of MCZ and 85% of SER after 24 h of treatment. Furthermore, this system eliminated a metabolite of SER (desmethylsertraline or DSER), and no differences between the elimination rates of DSER and SER were observed throughout the treatment period. It was also confirmed that the MnP-Tween 80 system caused a complete loss of growth inhibition by MCZ of the gram-positive bacterium *Bacillus subtilis* and reduced the growth inhibition by SER of the green alga *Pseudokirchneriella subcapitata* by 78% after 24 h of treatment. On the other hand, no appreciable elimination of MCZ and SER was obtained by treatment with laccase, MnP, or the laccase-mediator system with 1-hydroxybenzotriazole.

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Introduction

Numerous pharmaceuticals have been found in effluent from sewage treatment plants (STPs) and ultimately surface water. Although they are designed as bioactive molecules for clinical use and occur in the environment only in low concentrations, negative effects on non-target aquatic organisms are possible with the chronic low-dose exposure. Thus, pharmaceuticals are becoming recognized as emerging environmental contaminants (Santos et al., 2010; Queiroz et al., 2012; Backhaus and Karlsson, 2014).

Miconazole (MCZ) is an imidazole fungicide used for treatment of skin and vaginal fungal infections, and is also found in anti-dandruff shampoos. The antifungal effects of imidazole fungicides are due to inhibition of cytochrome P450 (CYP) isoenzyme 51 (CYP51/lanosterol-14α-demethylase) in fungi, which blocks the ergosterol biosynthesis essential for formation of the fungal cell membrane (Espinel-Ingroff, 1997; Joseph-Horne and Hollomon, 1997; Georgopapadakou, 1998). CYP51 is also found in many other species from bacteria to mammals, and it has been reported that MCZ inhibits human CYP51, which is required for the production of sterols important for germ cell development (Rozman et al., 2002; Zarn et al., 2003; Trösken et al., 2006). Furthermore, in mammals and fish, MCZ has been shown to inhibit CYP19 (aromatase), which converts androgens into estrogens and is therefore a key enzyme in the regulation of estrogens levels (Mason et al., 1985, 1987; Trösken et al., 2004; Lee et al., 2006; McKinlay et al., 2008). Thus, MCZ is suspected to potentially affect the endocrine systems of aquatic vertebrates and humans by inhibiting CYP19 and CYP51 involved in steroidogenesis. MCZ has been detected at the ng l^{-1} level in STP effluents in the UK and China (Roberts and Bersuder, 2006; Huang et al., 2010) and at concentrations ranging from 160 to 2069 ng g^{-1} (dry weight) in STP sludges in China, Sweden, and the U.S. (Huang et al., 2010; Lindberg et al., 2010; McClellan and Halden, 2010; Peng et al., 2012). Moreover, a recent study demonstrated that an average of 90% of MCZ in raw wastewater is adsorbed onto and persists in STP sludge (Peng et al., 2012). This suggests that the elimination of

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http://dx.doi.org/10.1016/j.ibiod.2015.02.017
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MCZ in STP is primarily attributable to adsorption of MCZ onto STP sludge, but not to microbial degradation of MCZ. As a consequence of its potential as an endocrine disruptor and its environmental persistence, MCZ nitrate is included on the List of Substances of Possible Concern presented by the Oslo and Paris (OSPAR) Commission (Roberts and Bersuder, 2006; OSPAR Commission, 2014).

The use of selective serotonin reuptake inhibitors (SSRIs) is rapidly increasing and these substances are among the most widely distributed antidepressants in the world. Sertraline (SER), a commonly prescribed SSRI antidepressant, has been shown to have adverse effects on a variety of non-target aquatic organisms, including algae, crustaceans, fish and frogs. Moreover, it has been reported that SER is more toxic than other common SSRIs (e.g., fluoxetine, fluboxamine, paroxetine, citalopram) in algae, crustaceans, and frogs (Henry et al., 2004; Richards and Cole, 2006; Christensen et al., 2007; Henry and Black, 2007; Johnson et al., 2007; Minagh et al., 2009). SER has been detected at ng l^{-1} concentrations (0.9–37.5 ng l^{-1}) in STP effluents and at concentrations ranging from 203 to 528 ng g^{-1} (dry weight) in anaerobically digested biosolid from STPs. In humans, SER is metabolized to desmethylsertraline (DSER), which has also been detected at concentrations in the range of 3.6–26.7 ng l^{-1} in STP effluents (Vasskog et al., 2006, 2008; Lajeunesse et al., 2008, 2012; Metcalfe et al., 2010; Schultz et al., 2010). Furthermore, DSER and SER have been found to accumulate in fish in municipal effluent-dominated streams (Brooks et al., 2005; Metcalfe et al., 2010; Schultz et al., 2010). Thus, SER has the potential to adversely affect aquatic organisms through chronic exposure.

Recently, ligninolytic enzymes, such as laccase, manganese peroxidase (MnP), lignin peroxidase, and versatile peroxidase, from lignin-degrading white-rot fungi have attracted significant research attention for their ability to eliminate endocrine disruptors (Tsutsumi et al., 2001; Suzuki et al., 2003; Tamagawa et al., 2005, 2007; Cabana et al., 2007; Sei et al., 2008; Mizuno et al., 2009), as well as pharmaceuticals and personal care products (Hata et al., 2010; Inoue et al., 2010; Marco-Urrea et al., 2010; Murugesan et al., 2010; Tran et al., 2010; Wen et al., 2010; Zhang and Geißen, 2010; Eibes et al., 2011; Suda et al., 2012; Weng et al., 2012; Touahar et al., 2014).

Laccase reduces molecular oxygen to water and simultaneously performs one-electron oxidation of various phenolic compounds. The substrate range of laccase is expanded by the addition of redox mediators such as 1-hydroxybenzotriazole (HBT); HBT radicals generated by laccase oxidation can oxidize nonphenolic compounds that are hardly oxidized or not oxidized at all by laccase alone (Fabbrini et al., 2002). On the other hand, MnP catalyzes the oxidation of Mn(II) to Mn(III) in the presence of H_2O_2. Organic acids such as malonate and oxalate chelate the generated Mn(III) and release Mn(III) from the manganese-binding site of MnP. The released Mn(III)-organic acid complex in turn can oxidize various phenolic compounds but cannot oxidize nonphenolic ones. It has been reported that Tween 80 containing the unsaturated fatty acid is able to extend the substrate range of MnP to nonphenolic lignin substructures (Bao et al., 1994; Jensen et al., 1996) and polycyclic aromatic hydrocarbons (Moen and Hammel, 1994; Bogan et al., 1996). Manganese peroxidase promotes lipid peroxidation of unsaturated fatty acid, possibly through Mn(III) chelates, thereby generating lipid peroxyl and other lipid radicals, which in turn can react to nonphenolic compounds as active oxidants (Mester and Tien, 2000; Aguiar et al., 2010).

In this study, we used four enzymatic treatments in an attempt to eliminate MCZ and SER (laccase, MnP, the laccase-HBT system, and the MnP-dependent lipid peroxidation system with Tween 80); results show that among these, the MnP-Tween 80 system is the most effective in achieving this goal.

Materials and methods

Chemicals

Miconazole nitrate and HBT were obtained from Wako Pure Chemical Industries (Osaka, Japan). Sertraline hydrochloride, oleic acid, linoleic acid, and lauric acid were purchased from Tokyo Kasei Kogyo (Tokyo, Japan). Tween 20 and Tween 80 (Surfact-Amps grade) were obtained from Thermo Scientific Pierce Protein Research Products (Rockford, IL) and DSER hydrochloride from Toronto Research Chemicals (Toronto, Canada). All other chemicals were extra pure grade, obtained from commercial sources, and used without further purification. The chemical structures of the target compounds, MCZ, SER, and DSER, are illustrated in Fig. 1.

Enzyme preparation and assay

Laccase from *Trametes versicolor* (IFO-6482) and MnP from *Phanerochaete chrysosporium* (ME-446) were prepared and partially purified according to our previous reports (Fujisawa et al., 2001; Tsutsumi et al., 2001). Laccase activity was determined by monitoring the oxidation of 2,6-dimethoxyphenol (DMP) to coerulignone. The reaction mixture contained 10^{-3} mol l^{-1} DMP and 5×10^{-2} mol l^{-1} malonate buffer (pH 4.5). Manganese peroxidase activity was determined in the same manner, except that the reaction mixture also contained 10^{-3} mol l^{-1} $MnSO_4$ and 2×10^{-4} mol l^{-1} H_2O_2. One katal (kat) of each enzyme activity was defined as the amount of enzyme producing 1.0 mol of coerulignone (49.6×10^3 mol^{-1} l cm^{-1} at 470 nm) from DMP per second.

Treatment of MCZ, SER, and DSER with ligninolytic enzymes

For treatment with laccase, the reaction mixture consisted of 10^{-4} mol l^{-1} MCZ nitrate or SER hydrochloride (10^{-2} mol l^{-1} stock solution in methanol), partially purified laccase (10 nkat ml^{-1}) and 5×10^{-2} mol l^{-1} malonate buffer (pH 4.5). The laccase-HBT system was used in the same manner, except that HBT (2×10^{-4} mol l^{-1}) was added to the reaction mixture for laccase treatment. The MnP reaction mixture consisted of 10^{-4} mol l^{-1} MCZ nitrate or SER hydrochloride, partially purified MnP (10 nkat ml^{-1}), 5×10^{-2} mol l^{-1} malonate buffer (pH 4.5), $MnSO_4$ (10^{-4} mol l^{-1}), and glucose (25×10^{-3} mol l^{-1}) plus glucose oxidase (4.2 nkat ml^{-1}; Wako Pure Chemical Industries) for the H_2O_2 supply. For the MnP-Tween 80 system, 0.1% (w/v) Tween 80 was added to the reaction mixture for

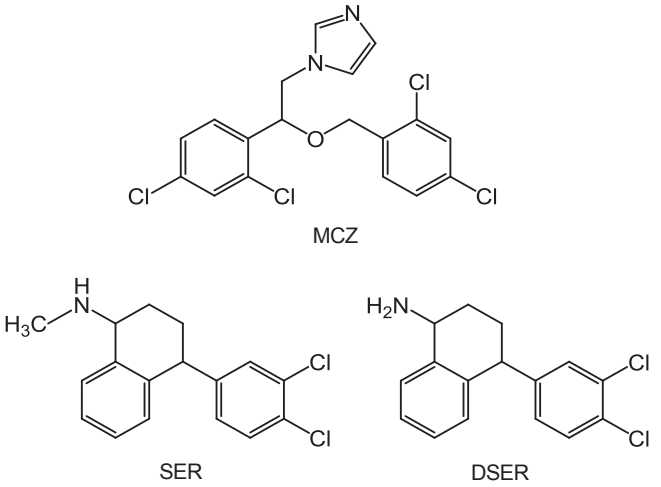

Fig. 1. Chemical structures of MCZ, SER, and DSER.

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