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Evaluation of reduction estrogenic activity in the combined treatment of landfill leachate and sanitary sewage

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ABSTRACT

Combined treatment of urban solid waste leachate and domestic sewage has emerged in Brazil and worldwide as an alternative to minimize the difficulties of treating landfill leachate. Landfill leachate is a complex matrix of organic and inorganic substances, recalcitrant material and xenobiotic organic compounds that are considered to be highly toxic, as well as endocrine disrupting compounds (EDC) that contribute to increased estrogenic activity. Although combined treatment is feasible for removal of organic matter, ammonia and toxicity, the efficiency of the removal of estrogenic substance is poorly investigated. In this study, the activated sludge process in batch and continuous modes was used for the combined treatment of 2% and 5% leachate/sewage mixtures. Estrogenic activity was reduced in reactors in both batch and continuous modes. The activated sludge process achieved average reductions of the estradiol equivalent (EQ-E2) values in the combined treatment of 94.5, 60.8 and 66.4% for the sanitary sewage, 2% mixture leachate in sewage and 5% mixture leachate in sewage reactors, respectively, in batch mode. For continuous reactors, 86%, 72%, and 65% of reduction of EQ-E2were achieved for sanitary sewage, 2 and 5% mixture with sanitary sewage. As for EQ-E2, the increase in the concentration of leachate in the sewage mixture was detrimental to the removal of estrogenic compounds during the biological process. Therefore, a limit on the % leachate/sewage mixtures should be established to achieve efficiency in the combined treatment of leachate in a Wastewater Treatment Plant (WWTP).

1. Introduction

Leachate is a liquid that results from the solubilization of organic and inorganic compounds due to the percolation of water from precipitation through the refuse mass (El-Fadel et al., 1997) and includes several toxic substances that directly affect human health and the environment when released without adequate treatment.

The National Policy on Solid Waste (NPSW) was established by Federal Law n. 12.305 in August 2010 and is considered a milestone for waste management in Brazil. One of the primary goals of this law is to close the open dumps and to perform appropriate disposal of MSW (Municipal Solid Waste) in landfills (Alfaia et al., 2017). Hence, new landfills are required that will treat the leachate generated properly.

In this scenario, combined treatment emerges as a suitable and less costly measure for leachate treatment because this type of treatment consists of mixing a portion of leachate into the sanitary sewage in a Wastewater Treatment Plant (WWTP). Therefore, after treatment, the effluent standards are sufficient for discharge (Verma and Kumar, 2016; Diamadopoulos et al., 1997).

The mixing ratios (on a volume basis) of landfill leachate in sanitary sewage are discussed in the literature. McBean et al. (1995) state that mixing ratios should not exceed 2%. For a mixing ratio of 10%, Diamadopoulos et al. (1997) found removals of 70–98% for BOD and 35–50% for NH_3—N. Nascentes (2013) evaluated mixing ratios of 0.5, 2, 3 and 5% and found a decrease in biodiversity in the biological treatment (activated sludge process) as the leachate concentration in the mixture increased, but the microbiological diversity and activity were quickly restored in the system. Yuan et al. (2016) evaluated landfill leachate co-treated with 14 municipal wastewaters at different mixing ratios and found that the 2.5% mixing ratio of leachate with wastewater improved the overall biological nutrient removal process of the system without

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compromising the COD removal efficiency. Brennan et al. (2017) found that the addition of intermediate leachate (produced in a landfill 2–10 year in age) at volumetric rates up to 4% did not significantly inhibit nitrification processes, whereas young leachate (produced in a landfill < 2 years in age) loaded at volumetric rates greater than 2% resulted in a significant decrease in nitrification. The co-treatment of old leachate (produced in a landfill >10 years in age) in municipal WWTPs may represent the most sustainable solution for on-going leachate treatment of the cases examined.

The combined treatment of leachate in domestic sewage has been evaluated regarding the performance of WWTP treatment processes on the removal of organic material and reduction in toxicity. However, the effect on endocrine disrupter compounds (EDCs) in the leachate in the combined treatment in WWTPs should be investigated.

EDCs are in different environmental matrices such as treated and raw sewage, landfill leachate, biological sludge, surface water and groundwater, among others (Behnisch et al., 2001; Bila and Dezotti, 2003). EDCs are substances with capacity to alter the endocrine system functions of humans and animals, affecting their growth and reproduction and the evolution of diseases such as cancer, fertility disorders, and abnormal sexual development (Sumpter and Johnson, 2005; Birkett and Lester, 2003; Coors et al., 2003).

Because of the varied composition of urban solid waste, leachate contains several EDCs of different chemical classes, such as polycyclic aromatic hydrocarbons (PAHs), phthalates, polychlorinated biphenyls (PCBs), biphenols (bisphenol A), alkyl phenols (nonylphenol and octylphenol), brominated flame retardants and natural and synthetic oestrogens (17β-estradiol, 17α-ethinylestradiol, estrone and estriol) (Asakura et al., 2004; Behnisch et al., 2001).

Among EDCs, natural and synthetic estrogens are substances with high estrogenic potential. Purdom et al. (1994) reported on the effects of estrogenic activity on fish exposed to effluents from WWTPs, and according to the authors, EE2 (17α-ethynylestradiol) concentrations of 0.1–0.5 ng/L are responsible for the induction of VTG (Vitellogenin, a protein that is used as a vertebrate biomarker of exposure to environmental estrogens, which stimulate elevated levels in males and females) production in male fish.

However, the determination of estrogenic activity and the compounds that act as endocrine disruptors in the leachate have received little attention. Therefore, detailed study of the behaviour of these compounds is necessary, to determine concentrations in the effluent and particularly, to study efficient treatment methods for their removal.

In vitro assays are tools widely used to evaluate estrogenic activity in different environmental matrices and combined with chromatographic analysis have been used to investigate the removal of estrogenic compounds in WWTPs.

The *in vitro* YES assay response is a result of the molecular cascade of events involved in receptor activation by estrogenic substances that bind to it and therefore provides an indication of the total estrogenic activity of a sample, and the response is directly dependent on the concentration of estrogenic substances (Zacharewski, 1997). In *vitro* assay to determine the estrogenic activity of the samples, in addition to measuring all estrogenic substances present in a sample, measure the interactions between these substances, whether synergistic or additive, which cannot be predicted only by quantification of all estrogenic substances in a sample.

In this scenario, the aim of the present study was to evaluate the estrogenic activity in the combined treatment of landfill urban solid waste leachate and sewage using the *in vitro* recombinant Yeast-Estrogen-Screen (YES) assay. Reduction efficiency of estrogenic activity in the biological treatment by the activated sludge process was evaluated.

2. Materials and methods

This work was carried out in experiments using reactors simulating the biological process of activated sludge, in bench scale, in batch and continuous mode. The aim of this work was to monitor the organic matter removal in global terms, by evaluation of the parameters COD, TOC and absorbance at 254 nm, the latter indicative of aromatic organic substances that absorb at 254 nm (APHA, 2012), which in the landfill leachate are represented by humic substances (Lima et al., 2017), as well as chromatographic analyzes (HPLC) and estrogenic activity to evaluate the estrogens selected in this study.

2.1. Chemicals

Standard solutions of 17β-estradiol, estriol, and 17α-ethynylestradiol were purchased from Sigma Aldrich. Ultra-pure water was obtained from a Milli-Q Biocell system (Millipore). Acetonitrile, ethyl acetate, and ethanol were obtained from Tedia, and the methanol and hydrochloric acid were obtained from Merck. All solvents used were HPLC/spectrum grade.

2.2. Landfill leachate

The landfill leachate samples were collected from the Seropedica landfill in October 2016 and May 2017. According to Costa et al. (2018), the Seropedica landfill was created in 2011 and receives approximately 9.000 t/day of urban solid waste from Rio de Janeiro City. This landfill is located in Seropedica Municipality that is 70 km from Rio de Janeiro City. The production of leachate is approximately 1.000 m^3 per day.

2.3. Effluents preparation

The preparation of effluents for the combined treatment was conducted with leachate mixed in different proportions with synthetic sewage. Synthetic sewage was produced according to methodology described in Fernandez Bou et al. (2015) using tap water, as presented in Table 1.

According to Cunha et al. (2016), some studies detected the presence of estrogens in domestic sewage in Brazil. Thus, in order for synthetic sewage to represent the quality of the sewage generated in Brazil, it was necessary to add estrogens.

Additionally, the synthetic sewage was spiked with 100 ng/L of each estrogen (17β-estradiol, estriol, and 17α-ethynyl estradiol) to obtain a minimum concentration of 100 ng/L of each compound, based on values of these compounds in domestic sewage, according to Cunha et al. (2016).

The determination of leachate proportions in sewage was based on information from previous studies on the efficiency of combined treatment in Brazil (Fernandez Bou et al., 2015; Mannarino et al., 2010). Therefore, the percentages of leachate in sewage were 2% and 5%.

Table 1
Composition of synthetic sewage (for COD of 625 ± 75 mg/L).

Compounds	Concentration (mg/L)
Casein peptone	360
Meat extract	250
Urea	100
Monobasic potassium phosphate	26
Sodium Chlorine	14
Calcium Chloride Dihydrate	8
Magnesium sulfate heptahydrate	4