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Review

Ecological risk of estrogenic endocrine disrupting chemicals in sewage plant effluent and reclaimed water

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ABSTRACT

The long-term ecological risk of micropollutants, especially endocrine disrupting chemicals (EDCs) has threatened reclaimed water quality. In this study, estrogenic activity and ecological risk of eight typical estrogenic EDCs in effluents from sewage plants were evaluated. The estrogenic activity analysis showed that steroidal estrogens had the highest estrogenic activity (ranged from 10^{-1} to 10^{3} ng-E2/L), phenolic compounds showed weaker estrogenic activity (mainly ranged from 10^{-3} to 10 ng-E2/L), and phthalate esters were negligible. The ecological risk of the estrogenic EDCs which was characterized by risk quotient ranged from 10^{-4} to 10^{3}, with an order in descending: steroids estrogens, phenolic compounds and phthalate esters. The eight estrogenic EDCs were scored and sorted based on the comparison of the estrogenic activity and the ecological risk, suggesting that 17α-ethynylestradiol (EE2), estrone (E1) and estradiol (E2) should be the priority EDCs to control in municipal sewage plants.

1. Introduction

Wastewater reclamation and reuse is an effective way to improve the utilization of water resources, which involves resolving the conflict of the water resources shortage, and mitigating the problem of water pollution (Janssens et al., 1997). Micropollutants, such as estrogenic endocrine disrupting chemicals (EDCs), in wastewater are considered to be harmful to human beings and aquatic organisms. Thus estrogenic EDCs in reclaimed water have received increasing interest and attention in recent years, and have been studied extensively in some developed countries (Laganà et al., 2004; Nelson et al., 2007; Sowers et al., 2009a; Zhang et al., 2012a).

Among the estrogenic EDCs that has been focused on (Birnbaum and Fenton, 2009; Saradha and Mathur, 2006) natural and synthetic estrogens are the most potent in *in vitro* (Cargouët et al., 2004; Gutendorf and Westendorf, 2001; Safe and Gaido, 1998) and *in vivo* studies (Sumpter and Jobling, 1995; Laws et al., 2000). The municipal wastewater effluents and pulp/paper industry discharges containing estrogens can derail the endocrine hormone in wild lives. Salmon, an example of endocrine disruption observed in fish species, has a high proportion of vitellogenin levels in the receiving water (Fernandez et al., 2007). The issue of estrogenic contaminants in wastewater has been recognized by scientists around the world, and the ability of wastewater treatment processes to mitigate current and future environmental risks from these compounds is being investigated by many researchers (Andersen et al., 2003; Drewes et al., 2005; Joss et al., 2004; Kirk et al., 2002; Shi et al., 2004; Zhang et al., 2012b, 2013). There has been proposed for EDCs quality specifications into wastewater effluents discharge standard in Britain (Butwell et al., 2005), it was intended for the estrone (E1), 17-β-estradiol (E2) and ethinyl estradiol (EE2) as the representative of estrogenic substances to limit discharge amount into the surrounding environment. However, the long term ecological risk of EDCs in municipal wastewater effluents and reclaimed water has been received continuous attentions.

The main objectives of this study were to evaluate the estrogenic activity and ecological risk of estrogenic EDCs in reclaimed water, and to propose precedent-controlled estrogenic EDCs during

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wastewater reuse accordingly. The estrogenic activity and ecological risk of estrogenic EDCs were characterized by estradiol equivalency (EEQ) and risk quotient (RQ), respectively, which both were calculated from measured environmental concentration (MEC) of EDCs. Therefore, the concentrations of the estrogenic EDCs in wastewater plant effluent were collected from major international journals during the past decade to calculate the estrogenic activity and ecological risk of estrogenic EDCs in reclaimed water.

2. Materials and methods

2.1. Data collection

Three main categories (totally eight kinds) of EDCs in the sewage plant effluents and reclaimed water were investigated. The first category steroidal estrogens contains E1, E2, estriol (E3) and EE2; the second category phenolic compounds contains nonylphenol (NP) and bisphenol A (BPA); the last category phthalate esters comprises dibutyl phthalate (DBP) and dibutyl phthalate (2-ethylhexyl) phthalate (DEHP).

The MEC and the estradiol equivalency factor (EEF) of the typical estrogenic EDCs in sewage plants effluents were collected mainly from the scientific papers published in the last decade, which could ensure the reliability of the data. In order to avoid data duplication, the data cited in the reviews were traced back to the original papers. The data were from: Aguayo et al., 2004; Andersen et al., 2003; Barber et al., 2007; Bertin et al., 2009; Bicchi et al., 2009; Braga et al., 2005; Baronti et al., 2000; Campbell et al., 2006; Cargouët et al., 2004; Clara et al., 2005; De Mes et al., 2005; Desbrow et al., 1998; Duong et al., 2010; Falconer et al., 2006; Fernandez et al., 2007; Fukazawa et al., 2002; Grover et al., 2011; Hansen, 2007; Hutchinson et al., 2000; Ingrand et al., 2003; Jiang et al., 2005; Johnson and Williams, 2004; Johnson et al., 2007; Jonkers et al., 2009; Kawahata et al., 2004; Körner et al., 2000; Kuch and Ballschmiter, 2001; Laganà et al., 2004; Leusch et al., 2006; Liscio et al., 2009; Liu et al., 2009; Matsui et al., 2000; Musolff et al., 2009; Nakada et al., 2004, 2007; Nakari, 2004; Nasu et al., 2001; Nelson et al., 2007; Nie et al., 2012; Onda et al., 2002, 2003; Park et al., 2009; Pawlowski et al., 2003, 2004; Petrovic et al., 2002, 2004; Pothitou and Voutsa, 2008; Roda et al., 2006; Rodriguez-Mozaz et al., 2004; Shao et al., 2005; Sheahan et al., 2002; Sowers et al., 2009a, 2009b; Stasinakis et al., 2008; Tan et al., 2007; Trenholm et al., 2006; Williams et al., 2003; Ying et al., 2002, 2009; Yu and Chu, 2009; Zhang and Zhou, 2008; Zhang et al., 2006; Zorita et al., 2009.

2.2. Analysis of estrogenic activity of EDCs

The estrogenic activity is represented by the estradiol equivalency (EEQ), which is calculated by the EEF and the MEC shown as Formula (1).

$$EEQ = EEF \times MEC \quad (1)$$

2.3. Analysis of ecological risk of EDCs

The RQ is used to evaluate the ecological risk of the typical estrogenic EDCs, and RQ relates to MEC and predicted no effect concentration (PNEC), shown as Formula (2).

$$RQ = MEC/PNEC \quad (2)$$

2.4. Data analysis methods

The values of EEQ and RQ varied with different MEC values. The distributions of EEQ and RQ were analyzed by the cumulative frequency, and the values of cumulative frequency were calculated by SPSS (Statistical Package from the Social Science) V13.0 statistics program. The cumulative frequency plots were drawn and the data that were out of reasonable range were discarded. Discrepancy were considered to be significant when $p < 0.05$ according to the Student's t-test.

3. Results

3.1. Estrogenic activity of typical estrogenic EDCs

3.1.1. Estrogenic activity of each estrogenic EDC

The estrogenic activity can be described as an interference caused by the environmental EDCs which interfere or damage the organisms' endocrine system in the ecosystem and the correlative system. The estrogenic activity' interference encompasses interference effects on reproduction and growth of the organisms, disorders on endocrine system and nervous system, and abnormality on immune function (Campbell et al., 2006; Crisp et al., 1998). For environmental and ecological systems, EDCs is mainly studied based on the biological individual cells, sub-organ or organs and tissue to reveal the influence mechanism for biological population, biotic community and system level in the ecological system.

Table 1
EEF of typical estrogenic EDCs.

Chemical	EEF			
	Sample number	Observed range	Median	95th percentile
E1	12	0.01–1.00	0.10	0.69
E2	15	1	1	1
E3	8	0.01–0.40	0.08	0.32
EE2	11	0.60–8.71	1.20	5.11
BPA	21	5.00×10^{-7}–0.05	5.00×10^{-5}	1.00×10^{-3}
NP	23	7.20×10^{-7}–0.01	6.93×10^{-5}	4.90×10^{-4}
DBP	8	1.80×10^{-8}–2.57×10^{-5}	6.80×10^{-7}	2.50×10^{-6}
DEHP	6	2.20×10^{-7}–1.30×10^{-5}	3.00×10^{-7}	1.30×10^{-5}

The EEFs of the eight estrogenic EDCs were shown in Table 1 and Fig. 1. The three categories EDCs were sorted in descending order of steroids estrogens, phenolic compounds and phthalate esters. The 95th percentiles of the EEFs were selected for the calculation of EEQs in this paper.

European Commission sets the concentration causing endocrine disrupting effects as 1 ng-E2/L, indicating that the substances with EEQ larger than 1 ng-E2/L would affect the endocrine systems of aquatic organisms in the receiving water (European Commission, 1996). The estrogenic activities of the eight estrogenic EDCs in sewage plant effluents and reclaimed water were shown in Fig. 2. The steroidal estrogens had the highest estrogenic activity among the three categories of estrogenic EDCs in the sewage plant effluents over the world, and most EEQ of the steroidal estrogens were larger than 1 ng-E2/L. Compared with the steroidal estrogens, the phenolic compounds showed weak estrogenic activity. However, 12% of EEQ of BPA and 15% of EEQ of NP were still higher than 1 ng-E2/L. EEQ of DBP and DEHP were below 1 ng-E2/L, thus the estrogenic activities of DBP and DEHP in the effluents were negligible.

3.1.2. Total estrogenic activity of estrogenic EDCs in different countries

In order to investigate the overall level of estrogenic activity in the sewage plants effluents, the total EEQ based on the EEQ of single estrogenic EDC were calculated by the Equation (3). EEQ of

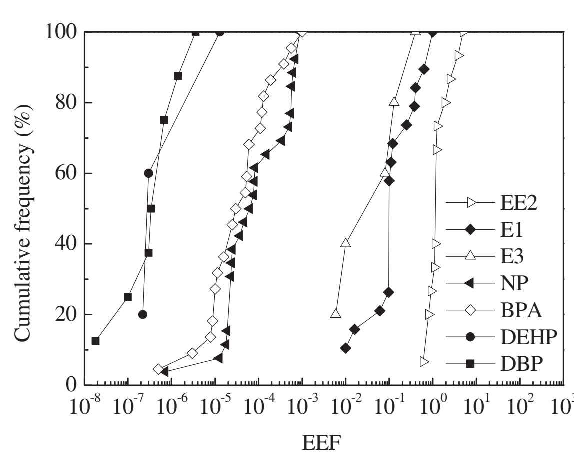

Fig. 1. EEF distribution of typical estrogenic EDCs.