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Technical Note

Removal of endocrine-disrupting chemicals and conventional pollutants in a continuous-operating activated sludge process integrated with ozonation for excess sludge reduction

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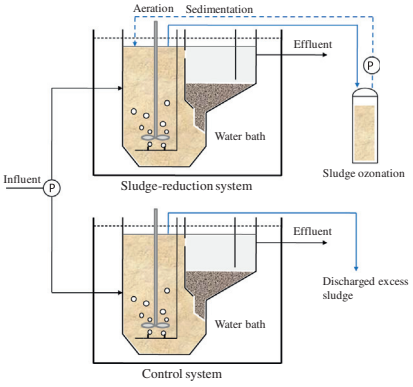
HIGHLIGHTS

- Sludge ozonation was applied to a continuous-operating activated sludge process.
- A zero yield of excess sludge was achieved at an ozone dose of 100 mg O_3 g^{-1} SS.
- Ozonation impacted insignificantly the removal of COD and nitrogen substances.
- Sludge ozonation contributed a little more removal of target EDCs than the control.
- Phosphorus recovery is necessary for the long-term sludge reduction by ozone.

GRAPHICAL ABSTRACT



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ABSTRACT

Sludge ozonation is considered as a promising technology to achieve a complete reduction of excess sludge, but as yet its effects on the removal of endocrine-disrupting chemicals (EDCs) and conventional pollutants (i.e., COD, N and P) in the activated sludge process are still unclear. In this study, two lab-scale continuous-operating activated sludge treatment systems were established: one was operated in conjunction with ozonation for excess sludge reduction, and the other was operated under normal conditions as control. The results indicate that an ozone dose of 100 mg O_3 g^{-1} SS led to a zero yield of excess sludge in the sludge-reduction system during a continuous-operating period of 45 d. Although ozonation gave a relatively lower specific oxygen uptake rate of activated sludge, it had little effect on the system's removal performance of COD and nitrogen substances. As a plus, sludge ozonation contributed a little more removal of target EDCs (estrone, 17β-estrodiol, estriol, 17α-ethinylestradiol, bisphenol A, and 4-nonylphenol). However, the total phosphorus removal declined notably due to its accumulation in the sludge-reduction system, which necessitates phosphorus recovery for the activated sludge process.

1. Introduction

In recent years, various excess sludge reduction technologies have been proposed which include mechanical, thermal, chemical, and biological methods (Wei et al., 2003). Sludge ozonation is considered as a promising technology because ozone can achieve a

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complete reduction of excess sludge at a cost comparable to or even lower than those of conventional methods (Yasui et al., 1996; Lee et al., 2005). However, this technology has some limitations (Chu et al., 2009), for example, an increased effluent COD concentration (Sakai et al., 1997; Huysmans et al., 2001), which implies that the sludge ozonation may deteriorate the effluent quality to some extent.

Public awareness of endocrine-disrupting chemicals (EDCs) has raised serious concerns due to their harmful effects on human health and aquatic ecosystem (Birkett and Lester, 2003). Especially, some estrogens could cause male fish feminization even at a trace concentration level (ng L^{-1}) in the aquatic environment (Purdom et al., 1994). Many studies (Ternes et al., 1999; Baronti et al., 2000) have proven that sewage treatment plant (STP) effluents are a crucial pollution source of EDCs to the environment. Among these, there are two main classes of EDCs in the STP effluents: steroid estrogens and phenolic compounds. The typical steroid estrogens include estrone (E1), 17β-estrodiol (E2), estriol (E3) and 17α-ethinylestradiol (EE2), among which EE2 is a synthetic estrogen used for contraceptive pills, whereas the others are natural estrogens mainly coming from urinary excretion. Phenolic compounds such as bisphenol A (BPA) and 4-nonylphenol (NP) are synthetic chemicals extensively used in household products such as textiles, detergents, and polymeric materials. Although BPA and NP only exhibit a low estrogenic activity, their detected concentration levels (μg L^{-1}) are usually much higher than those of steroid estrogens (ng L^{-1}) in the STPs (Liu et al., 2009; Nie et al., 2012).

In our previous study (Qiang et al., 2013), it was found that most target EDCs (more than 90% of estrogens and 60% of phenolic compounds) in the activated sludge could be removed during the sludge ozonation process at an ozone dose of 100 mg O_3 g^{-1} SS. However, it is still unclear if the sludge ozonation will affect the removal of EDCs in a continuous-operating activated sludge process, since ozonation may impact the microbial community and population (Yan et al., 2009). The microorganisms may directly influence the degradation of EDCs in the activated sludge process (Andersen et al., 2003). Therefore, it is necessary to investigate the removal of EDCs in a continuous-operating activated sludge process integrated with ozonation for excess sludge reduction, to evaluate the release risk of EDCs in the effluent.

In this study, two continuous-operating activated sludge treatment systems were established at lab scale and fed with synthetic wastewater. One was operated as the excess sludge reduction system, in which a part of sludge was ozonated and returned periodically to the biological oxidation tank, and the other was operated as the control system without sludge reduction. Hence, the removal efficiencies of target EDCs (including E1, E2, E3, EE2, BPA and NP) could be compared between the two treatment systems. Furthermore, the sludge-reduction efficacy and the biochemical performance of the activated sludge process (i.e., the removal of COD, N and P) were also evaluated.

2. Materials and methods

2.1. Sludge source and synthetic wastewater

The initial activated sludge was collected from the secondary sedimentation tank of an STP located in Beijing, China, which employed an anaerobic/anoxic/oxic biological treatment process. The collected sludge was cultivated under an aerobic condition with synthetic wastewater for 45 d before the experiments, to allow sludge acclimation and elimination of the background EDCs present in the sludge.

The synthetic wastewater, which was prepared daily, comprised the following components (mg L^{-1}): peptone (180), beef extract (120), $(NH_4)_2SO_4$ (100), K_3PO_4 (28), $MgSO_4$ (70), $CaCl_2$ (20), $FeCl_3$ (10), sodium acetate (32), and other trace elements. Its main quality parameters are shown in Table 1, which are comparable to those of typical municipal sewage except for the total nitrogen (TN). A higher TN concentration was prepared purposely in the synthetic wastewater to evaluate the potential impact of ozonation on the nitrification process of activated sludge.

The stock solution of target EDCs was prepared in methanol at 1000 mg L^{-1}, from which sub-stock solutions were prepared in Milli-Q water (resistivity >18.2 MΩ cm) that contained a mixture of estrogens (10 mg L^{-1}) and phenolic compounds (100 mg L^{-1}). After spiking, the final concentrations of target estrogens and phenolic compounds in the synthetic wastewater reached 1 and 10 μg L^{-1}, respectively. The chemical structures of target EDCs are shown in Fig. SM-1 in Supplementary Material (SM).

2.2. Activated sludge treatment systems

Two continuous-operating activated sludge treatment systems (i.e., sludge-reduction and control) were established in our laboratory and their photo is shown in Fig. SM-2. Each system consisted of an aeration tank and a sedimentation tank, with both made of stainless steel. Moderate stirring was performed in the aeration tank to mix the influent and the activated sludge. The two systems had the same wastewater treatment capabilities, and their volumes and operational conditions are listed in Table 1. For the control system, 250 mL of activated sludge was discharged daily; whereas for the sludge-reduction system, 400 mL of activated sludge was collected and ozonated via batch mode daily. An ozone generator (CF-G-3-010G, Guolin) was utilized to produce ozone-containing gas, whose flow rate was controlled at 1.5 L min^{-1} during the sludge ozonation. The gaseous ozone concentration was determined to be about 20 mg L^{-1} and an ozone dose of 100 mg O_3 g^{-1} SS was adopted. The ozonated sludge was continuously returned to the aeration tank by using a peristaltic pump, and no excess sludge was discharged during the entire operating period.

The synthetic wastewater was synchronously fed to the two treatment systems by using one peristaltic pump, which ensured an identical flow rate of the two influents. The entire operating period lasted for 60 d, which could be divided into two stages. In the first stage (0–15 d), the two treatment systems adopted the same operating strategy (i.e., the conventional activated sludge process) to examine their removal efficacy for conventional pollutants. In the second stage (16–60 d), sludge ozonation was applied

Table 1
Operation parameters and influent characteristics of the sludge-reduction and control systems.

Items	Sludge-reduction	Control
Operation parameters		
Bioreactor volume (L)	5.2	5.2
Settling tank volume (L)	2.6	2.6
MLSS (mg L^{-1})	1756 ± 90	1690 ± 50
HRT (h)	15	15
SRT (d)	N/A[a]	20
DO (mg L^{-1})	3.2–4.8	3.0–5.1
pH	4.9–7.1	5.2–6.9
Influent characteristics		
COD (mg L^{-1})	363 ± 5	
TN (mg L^{-1})	69 ± 2	
NH_3–N (mg L^{-1})	21.2 ± 1.2	
TP (mg L^{-1})	9.6 ± 1.8	
pH	~6.2	

[a] Not applicable, because there was no excess sludge discharged from the sludge-reduction system.