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Low-temperature anaerobic treatment of low-strength pentachlorophenol-bearing wastewater

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HIGHLIGHTS

- Anaerobic reactors for the treatment of low-strength wastewaters at low temperature.
- PCP decreases the COD removal and caused an irreversible inhibition on methanogenesis.
- Inhibitory effect was modelled by modified pseudo-Monod and Roediger models.
- EGSB reactor. A solution for PCP-containing wastewaters treatment at low temperature.

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ABSTRACT

The anaerobic treatment of low-strength wastewater bearing pentachlorophenol (PCP) at psychro-mesophilic temperatures has been investigated in an expanded granular sludge bed reactor. Using an upward flow rate of 4 m h^{-1}, a complete removal of PCP, as well as COD removal and methanization efficiencies higher than 75% and 50%, respectively, were achieved. Methanogenesis and COD consumption were slightly affected by changes in loading rate, temperature (17–28 °C) and inlet concentrations of urea and oils. Pentachlorophenol caused an irreversible inhibitory effect over both acetoclastic and hydrogenotrophic methanogens, being the later more resistant to the toxic effect of pentachlorophenol. An auto-inhibition phenomenon was observed at PCP concentrations higher than 10 mg L^{-1}, which was accurately predicted by a Haldane-like model. The inhibitory effect of PCP over the COD consumption and methane production was modelled by modified pseudo-Monod and Roediger models, respectively.

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1. Introduction

Pentachlorophenol (PCP) is a biocide used as wood preservative. It was extensively used as a wide spectrum pesticide until its prohibition in 2004 by the Rotterdam Convention. Large quantities of PCP are also produced from ECF-type paper pulp bleaching, the most commonly used nowadays (Savant et al., 2006). Due to its toxic character, PCP is of environmental concern since it acts on a variety of organisms as a potent inhibitor of the oxidative phosphorylation. It disrupts the proton gradient across membranes, interfering with energy transduction of cells (Chen et al., 2008). Moreover, its carcinogenic potential has been recently demonstrated (Cooper and Jones, 2008). The use of this chemical over decades, its resistance to biodegradation, its potential bioaccumulation and biomagnification (Letcher et al., 2009) have led to a wide spread in the environment. PCP has been identified as a priority hazardous substance by the European Community. The environmental quality standards for surface water established by the Directive 2000/60/EC limit the average and maximum allowable concentrations to 0.4 and a 1.0 μg L^{-1}, respectively.

Some tropical countries have continued using PCP as pesticide and component of several agrochemicals in agricultural areas, which is the main cause of its presence in surface and underground water, where concentrations up to 20 mg L^{-1} have been detected in some cases (Downs et al., 1999). Although its contribution as environmental pollutant has declined, PCP still appears in water reservoirs even in countries where this chemical has been banned for several years (Damianovic et al., 2009), reaching concentrations up to 0.36 μg L^{-1} (Gasperi et al., 2008). The presence of PCP in urban areas is thought to be caused through its leakage to the municipal sewer system as a result of accidental spills and dumps. Recent studies have identified the contribution of PCP to contaminant loads into domestic wastewater, reaching concentrations between 0.02 and 0.05 μg L^{-1} in domestic greywater (Nielsen et al., 2005). Treatment by conventional systems appears to be insufficient, since pesticides have been detected in resulting effluents. Thus, highly-efficient removal from wastewater is needed to prevent potential human health risks associated with the ingestion over long periods.

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Table 1
Experiences on anaerobic biological treatment of PCP in lab scale reactors.

Reactor[a]	PCPLR (mg PCP L^{-1} d^{-1})	PCP (mg L^{-1})	HRT (d)	OLR (g COD L^{-1} d^{-1})	T (°C)	Co-substrate[b]	Chlorophenols detected	Refs.
UASB	97	60	NS	16.2	28	Met, Ace, Pro	NS	Wu et al. (1993)
UASB	0.3	1.0	2.0–3.0	0.4	35	Ph	DCP	Duff et al. (1995)
UASB-b	32	NS	0.2–1.0		NS	Lac	345TCP	Christiansen and Ahring (1996)
GAC-FBBR	0.4	1333	9.3	6.3	35	Eth	3CP, 4CP	Khodadoust et al. (1997)
FBBR	23	1.9	0.1	0.2	35	Glu or But	4CP, 2CP	Mohn et al. (1999)
FBBR	218	100	NS	NS	35	Eth	4CP, 3CP, 34DCP, 35DCP, Ph	Koran et al. (2001)
UASB	62	NS	1.2	0.9	35	Suc, But, Eth	345TCP, 35DCP, 3CP	Tartakovsky et al. (2001)
Hybrid reactor	16.8	21	1.3	6.9	32–37	Met, Ace, Pro, But	NS	Montenegro et al. (2001)
ASBR	5	5000	NS	5.0	28	Suc	NS	Ye and Shen (2004)
FFBR	313	364	1.2–1.5	4.5–5.6	35	Suc, But, Eth, YE	3CP	Lanthier et al. (2005)
UASB	181	152.0	1.0	12.0	28	Suc	NS	Shen et al. (2005, 2006))
HAIB	2–13	2–13	1.0	1.15	30	Glu, Ace, For, Eth	NS	Baraldi et al. (2008)
ASBR	17.7	4.4	2.0	NS	24–32	Suc	345TCP	Mun et al. (2008)
HAIB	4.1	8.0	0.7	1.7	30	Glu, Ace, For, Eth	DCP	Damianovic et al. (2009)

NS: Not specified.
[a] UASB-b: UASB reactor with biofilms; GAC-FBBR: FBBR with granular activated carbon; HAIB: Horizontal-flow anaerobic immobilized biomass reactor; ASBR: Anaerobic sequencing batch reactor.
[b] Ace: acetate; But: butyrate; Eth: ethanol; For: formiate; Glu: glucose; Lac: lactate; Met: methanol; Pep: peptone; Ph: phenol; Pro: propionate; Suc: sucrose; YE: yeast extract.

Chlorophenols have been commonly removed from effluents by adsorption, which transfers the problem to the spent adsorbent which becomes a hazardous waste. On the opposite, chemical and biological destructive methods allow the mineralization of chlorophenols. Although chemical oxidation methods are faster than biological ones, the potential generation of byproducts eventually more toxic than the starting pollutants can be a serious problem. The toxic effect of PCP in biological reactors leads to persistent-inherent and persistent-readily biodegradability for aerobic and anaerobic biological systems, respectively. Thus, the combination of both chemical and biological processes has been proposed to achieve high removal efficiencies (Essam et al., 2007).

Several works have reported on the anaerobic treatment of wastewaters containing chlorinated compounds by different technologies (Farhadian et al., 2008), including anaerobic bioreactors with immobilized biomass (Khodadoust et al., 1997; Lanthier et al., 2005; Montenegro et al., 2001), fluidized bed biofilm reactors (FBBR) (Mun et al., 2008), sequencing batch reactors (SBR) (Mun et al., 2008), up-flow anaerobic sludge blanket reactors (UASB) (Duff et al., 1995; Shen et al., 2005; Wu et al., 1993), and membrane anaerobic bioreactors have been recently presented as an effective technology to treat toxic compounds (Singhania et al., 2012). Table 1 shows the experimental conditions used for the treatment of PCP in lab scale anaerobic reactors and the byproducts detected in the resulting effluents. Most of the works dealing with anaerobic PCP removal have shown that low PCP concentrations (0.75–11.3 μM) inhibit methanogenesis completely (Duff et al., 1995; Wu et al., 1993). Juteau et al. (1995) reported 99% PCP removal in a fixed-film biological reactor (FFBR) with a PCP load of 60 μM d^{-1}. The major intermediates were 3,4-dichlorophenol (3,4-DCP) and 3-chlorophenol (3-CP) and no significant 3-CP degradation was observed. Damianovic et al. (2009) treated efficiently PCP loads up to 15.6 μM PCP d^{-1} in a horizontal-flow anaerobic immobilized biomass reactor. Beaudet et al. (1997) achieved complete dechlorination of PCP in an anaerobic FFBR fed with up to 68 μM d^{-1} of PCP extracted from contaminated wood chips. Among the high-performance anaerobic systems, UASB reactors have shown a better ability for treating higher PCP loads than FF bioreactors (Duff et al., 1995).

Under anaerobic conditions PCP is biodegraded through reductive dechlorination, leading to less-chlorinated chlorophenols and, in some cases, to complete dechlorination, which facilitates subsequent biodegradation (Wu et al., 1993). PCP anaerobic conversion has been mostly carried out under mesophilic conditions, which require around 30% of the energy associated to the biogas generated. However, low and medium strength wastewaters are commonly discharged at low ambient temperatures, including municipal wastewater and a broad variety of industrial wastewater. The treatment at low-temperature by psychrophilic anaerobic bioreactors has recently proved to be feasible for a range of wastewater categories, including the removal of trichloroethylene (Siggins et al., 2011), trichlorophenols (Collins et al., 2005) and nutrients (Ma et al., 2013). These emerging technologies represent a breakthrough for the management of highly polluted wastewaters. The high-performance anaerobic systems, like expanded granular sludge bed (EGSB) reactor, are promising potential solutions where the contact between the biomass and wastewater is enhanced. The high upflow velocity that can be applied in EGSB (4–10 m h^{-1}) provokes effective mixing, allowing improved efficiency (Puyol et al., 2009).

Wastewater treatment by EGSB reactors at low temperatures seems to be particularly promising for regions in which sewage temperatures do not drop below 15 °C (Lettinga et al., 2001) like in subtropical regions, including the Mediterranean countries. Thus, psychrophilic anaerobic digestion represents an attractive alternative and economically sound option for sustainable wastewater management. However a more in-depth knowledge of the response of low-temperature anaerobic systems to highly toxic micropollutants is needed. In this work, the anaerobic decontamination of synthetic wastewater bearing PCP at low temperature by an EGSB reactor is studied with the aim of analyzing the inhibition of methanogenesis caused by PCP which can provide useful information for design.

2. Methods

2.1. Wastewater composition

Synthetic wastewater was prepared by adding the following components (mg L^{-1}): peptone (17.4), yeast extract (52.2), milk

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