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Role of nitrification in the biodegradation of selected artificial sweetening agents in biological wastewater treatment process

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HIGHLIGHTS

- Role of nitrification in the biodegradation of ASs was first examined.
- Removal efficiency of ASs was enhanced under high initial NH_4-N concentrations.
- There was a relationship between nitrification rate and biodegradation rate of ASs.
- Ammonia monooxygenase played a key role in biodegradation of acesulfame and sucralose.

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ABSTRACT

The biodegradation of the six artificial sweetening agents including acesulfame (ACE), aspartame (ASP), cyclamate (CYC), neohesperidindihydrochalcone (NHDC), saccharin (SAC), and sucralose (SUC) by nitrifying activated sludge was first examined. Experimental results showed that ASP and NHDC were the most easily degradable compounds even in the control tests. CYC and SAC were efficiently biodegraded by the nitrifying activated sludge, whereas ACE and SUC were poorly removed. However, the biodegradation efficiencies of the ASs were increased with the increase in initial ammonium concentrations in the bioreactors. The association between nitrification and co-metabolic degradation was investigated and a linear relationship between nitrification rate and co-metabolic biodegradation rate was observed for the target artificial sweeteners (ASs). The contribution of heterotrophic microorganisms and autotrophic ammonia oxidizers in biodegradation of the ASs was elucidated, of which autotrophic ammonia oxidizers played an important role in the biodegradation of the ASs, particularly with regards to ACE and SUC.

1. Introduction

The occurrence and fate of emerging trace organic contaminants (EOCs) such as pharmaceuticals and personal care products (PPCPs) and endocrine disrupting chemicals (EDCs) in natural and engineered systems like wastewater treatment plants (WWTPs) has been increasingly gaining attention during the last decades (Hai et al., 2011; Huggett and Stoddard, 2011; Tadkaew et al., 2011; Wijekoon et al., 2013; Tran et al., 2014a). Similar to PPCPs, it has been recently found that ASs are ubiquitously present in a variety of environmental compartments, such as wastewater, surface waters, groundwater, and drinking water sources (Buerge et al., 2009; Scheurer et al., 2009, 2011; Oppenheimer et al., 2011; Van Stempvoort et al., 2011a; Lange et al., 2012; Tran et al., 2014b). Naturally, ASs end up in the environment through human consumption of low-calorie beverage, food and other products, in which ASs are used as sugar substitutes in remarkable amounts (Kroger et al., 2006). Consumed ASs are excreted through urine or faeces (Lange et al., 2012). The widespread occurrence of ASs in surface water systems is due to both their incomplete metabolism in the human bodies and their persistence in the environment (Buerge et al., 2009; Lange et al., 2012; Tran et al., 2013a). The concentrations levels of ASs in aquatic environment varied significantly from a few tens nanogram per Litre to a few tens microgram per Litre, depending upon environmental compartments and type of sweetener (Buerge et al., 2009; Tran et al., 2014b). For example, Buerge et al. (2009) reported that the concentrations of ASs in untreated wastewater samples ranged 12–43 μg/L for acesulfame, 10–65 μg/L for cyclamate, 3.0–18 μg/L for saccharin, and 2.0–9.1 μg/L for sucralose. In another study, Buerge et al. (2011) also released that saccharin was present in liquid manure at

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Table 1
The physicochemical properties of the target ASs.

Target AS	Molecular weight (g/mol)	log K_{ow}	pK_a	Henry's law constant (atm m^3/mol)
ACE	162.99	−1.33	2.0	9.6×10^{-9}
ASP	294.12	0.07	3.2/8.7	2.5×10^{-18}
CYC	178.05	−1.61	1.71	1.7×10^{-8}
SAC	182.99	0.91	2.2	1.2×10^{-9}
SUC	396.01	−1.0	11.8	4×10^{-19}
NHDC	612.2	0.75	9.7	6.3×10^{-17}

concentrations up to 12 mg/L, where it was stable during 2 months of storage. Recently, ASs are frequently found in most surface water (Buerge et al., 2009; Scheurer et al., 2009; Lange et al., 2012), groundwater (Buerge et al., 2009, 2011; Scheurer et al., 2009; Van Stempvoort et al., 2011a,b, 2013), and even in several tap water samples at concentrations up to µg/L levels (Buerge et al., 2009; Mawhinney et al., 2011). Numerous studies have been made to elucidate the bioaccumulation and ecotoxicological effects of ASs. However, to date, it has been still controversial on the toxicity of ASs to human health as well as aquatic organisms (John et al., 2000; Wiklund et al., 2012; Zhang et al., 2012). It has been thought that ASs such as sucralose do not have any effect on metabolism since the ASs are used in such small amounts that they do not increase calorie intake (Huggett and Stoddard, 2011; Tollefsen et al., 2012; Wiklund et al., 2012). Nevertheless, recent findings from the animal-based studies suggested that sucralose may be doing more than just making foods and drink taste sweeter (Zhang et al., 2012). For example, Zhang et al. (2012) demonstrated that sucralose affects the glycemic and insulin responses to an oral glucose load in obese people who do not normally consume non-nutritive sweeteners. They also found that when receptors in the gut are activated by ASs, the adsorption of glucose also increases. Although the toxicity of ASs to human health is relatively unknown at trace levels so far, continuous discharge and simultaneously chronic exposure to ASs and PPCPs/EDCs may increasingly pose a risk to human health. It is therefore required to remove ASs from wastewater and water to protect human health and aquatic environment, particularly with regards to the enhancement of public acceptance in using reclaimed water for water supply. To date, it is no surprise that there are a few studies emphasized on the removal of ASs in wastewater/water processes. Particularly, there is little information on the role of autotrophic ammonia oxidizers in the biodegradation of ASs. Hence, the objective of this study was to examine the biodegradation of the selected ASs by nitrifying activated sludge. Meanwhile, the relationship between nitrification and co-metabolic biodegradation of ASs was also elucidated.

2. Methods

2.1. Target ASs, chemical reagents, and solvents

The target ASs investigated in this study were acesulfame (ACE), aspartame (ASP), cyclamate (CYC), neohesperidindihydrochalcone

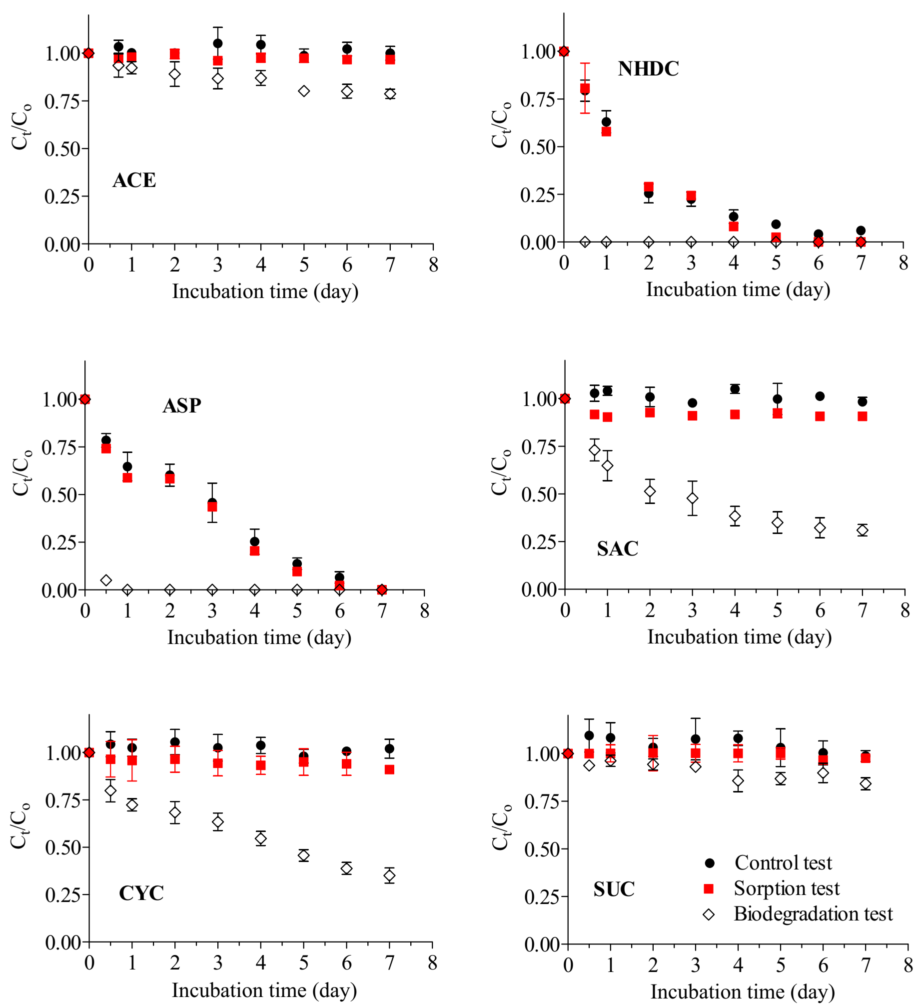

Fig. 1. Changes in the concentrations of target ASs with incubation time for the different kinds of experiments (control, sorption, and biodegradation).