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Emission of artificial sweeteners, select pharmaceuticals, and personal care products through sewage sludge from wastewater treatment plants in Korea

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ABSTRACT

Concern over the occurrence of artificial sweeteners (ASWs) as well as pharmaceuticals and personal care products (PPCPs) in the environment is growing, due to their high use and potential adverse effects on non-target organisms. The data for this study are drawn from a nationwide survey of ASWs in sewage sludge from 40 representative wastewater treatment plants (WWTPs) that receive domestic ($WWTP_D$), industrial ($WWTP_I$), or mixed (domestic plus industrial; $WWTP_M$) wastewaters in Korea. Five ASWs (concentrations ranged from 7.08 to 5220 ng/g dry weight [dw]) and ten PPCPs (4.95–6930 ng/g dw) were determined in sludge. Aspartame (concentrations ranged from 28.4 to 5220 ng/g dw) was determined for the first time in sewage sludge. The median concentrations of ASWs and PPCPs in sludge from domestic WWTPs were 0.8–2.5 and 1.0–3.4 times, respectively, the concentrations found in WWTPs that receive combined domestic and industrial wastewaters. Among the five ASWs analyzed, the median environmental emission rates of aspartame through domestic WWTPs (both sludge and effluent discharges combined) were calculated to be 417 μg/capita/day, followed by sucralose (117 μg/capita/day), acesulfame (90 μg/capita/day), and saccharin (66 μg/capita/day). The per-capita emission rates of select PPCPs, such as antimicrobials (triclocarban: 158 μg/capita/day) and analgesics (acetaminophen: 59 μg/capita/day), were an order of magnitude higher than those calculated for antimycotic (miconazole) and anthelmintic (thiabendazole) drugs analyzed in this study. Multiple linear regression analysis of measured concentrations of ASWs and PPCPs in sludge revealed that several WWTP parameters, such as treatment capacity, population-served, sludge production rate, and hydraulic retention time could influence the concentrations found in sludge.

1. Introduction

Artificial sweeteners (ASWs) are used as sugar substitutes in substantial quantities in foods, drinks, drugs, and hygiene products (Weihrauch and Diehl, 2004). Four ASWs, acesulfame, aspartame, saccharin, and sucralose are regarded safe as food additives in the USA (FDA, 2006), whereas the European Union additionally approved cyclamate (EU, 1994) (Fig. 1). In Korea, saccharin, acesulfame, aspartame, and sucralose were reported in children's snacks at concentrations of 0.41–197 mg/kg (Lee et al., 2011). The global consumption of acesulfame, cyclamate, and saccharin was 4000, 47,000, and 37,000 tons in 2005, respectively (ISO, 2008). The annual sale of sucralose in the USA in 2004 was >$170 million, and its estimated loading to the wastewater treatment plants (WWTPs) was over 10 million kg (Mead et al., 2009; Soh et al., 2011). In Korea, saccharin and aspartame were among the top 100 food additives in 2011, with annual production volumes of 348 and 838 tons, respectively (KFDA, 2012). Following ingestion, acesulfame, cyclamate, saccharin, and sucralose excrete from human bodies at rates of >90% of the ingested dose (Mayer and Kemper, 1991; Renwick, 1985) (EC, 2000); nevertheless, aspartame is almost completely metabolized in humans (Soffritti et al., 2006, 2010) (Fig. S1).

The U.S. Food and Drug Administration (FDA) banned cyclamate in 1970 from all dietary foods due to its potential to induce carcinogenesis in experimental animals (TPS, 1970). Chronic exposure to saccharin results in induction of bladder cancer in rats (Schoenig et al., 1985; Weihrauch and Diehl, 2004); aspartame was shown to exhibit carcinogenic effects in rats, primarily through its metabolic products (Soffritti et al., 2006). In aquatic organisms, sucralose exposure results in an increase in swimming speed in *Daphnia magna* (Wiklund et al., 2012)

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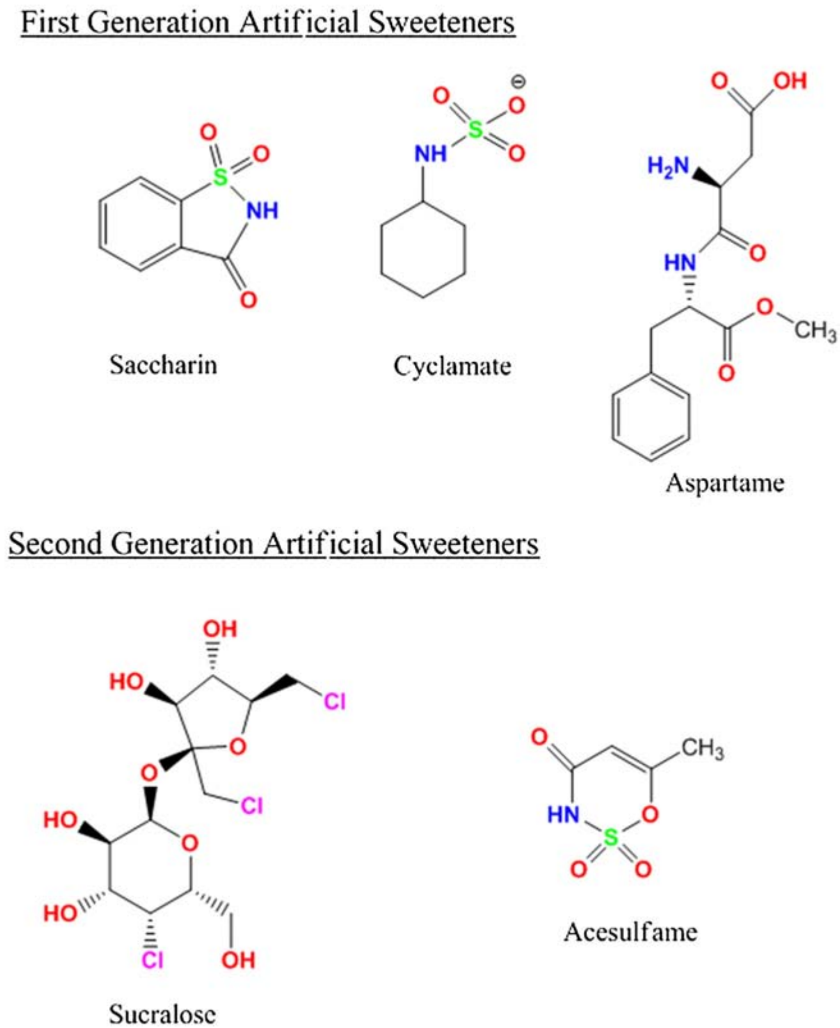

Fig. 1. Chemical structures of select artificial sweeteners analyzed in this study.

and an increase in the intake of food by a marine copepod, *Calanus glacialis* (Hjorth et al., 2010).

WWTPs are a major source of environmental emission of ASWs as well as pharmaceuticals and personal care products (PPCPs). Studies have shown that saccharin and cyclamate were efficiently removed (>90%) in WWTPs in Germany and Switzerland; however, acesulfame and sucralose were not removed efficiently (negative removal of 42%) (Buerge et al., 2009; Scheurer et al., 2009). ASWs and PPCPs were found in the effluent of WWTPs at concentrations of μg/L (Buerge et al., 2009; Lunden et al., 2008a,b; Scheurer et al., 2009). Several ASWs and PPCPs were found at μg/L concentrations in lake water (Buerge et al., 2009), river water (Scheurer et al., 2009), and drinking water systems (Mawhinney et al., 2011) in Germany, Switzerland, and the USA. Three studies, thus far, have reported the occurrence of ASWs in sludge from WWTPs. Buerge et al. (2011) and Ordonez et al. (2013) determined sucralose, acesulfame, saccharin, and cyclamate in sludge from WWTPs in Switzerland and Spain; and Lunden et al. (2008a,b) reported sucralose in sludge from two WWTPs in Sweden.

This is the first nationwide study to report the occurrence of ASWs in sludge from WWTPs in Korea. Five ASWs (sucralose, aspartame, saccharin, acesulfame, and cyclamate) and ten PPCPs (comprising two PCPs [triclocarban and oxybenzone], three analgesics [acetaminophen, ketoprofen, and codeine], three antibiotics [lincomycin, clindamycin, and trimethoprim], one antimycotic [miconazole], and one anthelmintic [thiabendazole]) were determined in sludge from 40 WWTPs that receive industrial and domestic wastewater. The rates of environmental emission of target ASWs and PPCPs through the discharge of sludge and wastewater effluent were calculated. The emission rates were estimated based on the concentrations determined in sludge and the concentrations predicted for effluents on the basis of reported sludge–water partition coefficients of target chemicals. Finally, factors that affect concentrations of ASWs in sludge were investigated by multiple linear regression analysis of WWTP parameters, such as treatment capacity, population-served, sludge production rate, and hydraulic retention time.

2. Materials and methods

2.1. Reagents and chemicals

Analytical standards of saccharin, sucralose, trimethoprim, trimethoprim-D_9, oxybenzone, oxybenzone-D_5, ketoprofen, acetaminophen, clindamycin hydrochloride, miconazole nitrate, lincomycin hydrochloride monohydrate, and thiabendazole were purchased from Sigma-Aldrich (St. Louis, MO, USA). Acesulfame potassium, cyclamate, and aspartame were purchased from Supelco (Bellefonte, PA, USA). Saccharin-$^{13}C_6$, sucralose-D_6, acesulfame-D_4, and aspartame-D_5 were purchased from Santa Cruz Biotechnology, Inc. (Dallas, TX, USA). Triclocarban and