



Mass loading of typical artificial sweeteners in a pig farm and their dissipation and uptake by plants in neighboring farmland



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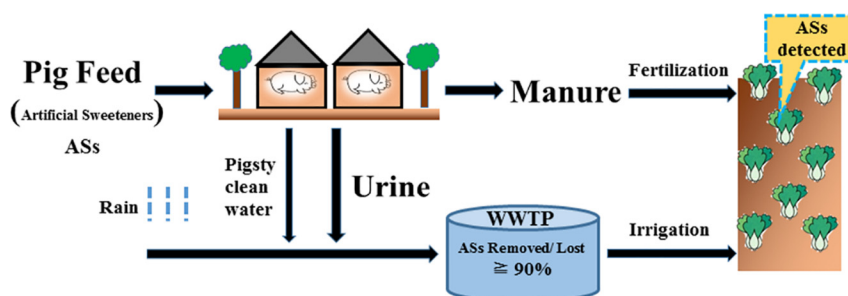
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HIGHLIGHTS

- Artificial sweeteners (ASs) were widely detected in pig feed and manure.
- ASs can enter soil via application of treated pig manure and urine in huge quantities.
- Plants (vegetables) can take up ASs markedly when the ASs concentrations were high in soil.
- Dissipation of ASs in field soil were quick with half-lives of several days.

GRAPHICAL ABSTRACT



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ABSTRACT

Pig farm is an important potential source for artificial sweeteners (ASs) in the environment due to their wide use as additives in pig feed. The objective of this study was to evaluate the fate of typical ASs in pig farm and neighboring farmland. For this purpose, the levels of four typical artificial ASs, i.e. saccharin (SAC), cyclamate (CYC), acesulfame (ACE) and sucralose (SUC), in pig feed and manure from a pig farm and water samples from an on-farm wastewater treatment plant (WWTP) in Tianjin, China were measured and the mass loadings and removal efficiencies were assessed. Moreover, the levels of ASs in different layers of soil and vegetables in neighboring farmland that received manure fertilizers and wastewater from the farm were consecutively monitored for 60–80 days. The SAC, CYC and ACE were widely determined in all kinds of the samples, while SUC was only found in few soil samples. The mass loadings of the ASs in pig feed were estimated up to 311 kg/year for SAC, 59.1 kg/year for CYC, and 17.1 kg/year for ACE, respectively. The fractions of the total mass of ASs excreted via manure were estimated to be 36.0% for SAC, 59.4% for CYC, and 36.7% for ACE as compared to those in pig feed. High removal efficiencies (>90%) of ASs in the on-farm WWTP was achieved. In greenhouse soils, CYC, SAC, ACE, and SUC were degraded quickly, with half-lives of 4.3–5.9 d, 2.7–4.2 d, 8.4–12.3 d, and 7.3–10.8 d, respectively. Lower levels of ASs were found in deeper soil layer (20–30 cm). The ASs were considerably absorbed by plants when the ASs' concentrations were high in soil. This study presents the first comprehensive overview of ASs fate from a pig farm to the neighboring agricultural ecosystem.

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1. Introduction

Artificial sweeteners (ASs) are widely used around the world as sugar substitutes in food additives, beverages, sanitary products, pharmaceuticals and even animal feeds (Kroger et al., 2006; Kokotou et al., 2012; Lange et al., 2012). Their sweetness is much higher than that of table sugar, and they provide no or negligible energy because they are not metabolized like sugars or only fermented slightly by the mouth microflora (Strubig, 1988; Imfeld, 1993). Acceptable daily intake values for ASs (Table 1) were recommended due to their potential health risks based on comprehensive toxicological tests (Kroger et al., 2006; Lubick, 2008; Soh et al., 2011; Qin, 2012; Wiklund et al., 2012; Pepino et al., 2013; Suez et al., 2014). For instances, some ASs, especially saccharin (SAC), can induce glucose intolerance and increase type 2 diabetes risk by altering the composition and function of mice gut microbiota (Suez et al., 2014). Sucralose (SUC) was claimed to be able to cause side effects such as increasing adsorption of glucose by triggering gut receptors (Pepino et al., 2013). A positive association between the use of ASs and risk of bladder cancer in males had been reported in the early years (Howe et al., 1977), but this conclusion had been challenged (Hoover and Strasser, 1980; Weihrauch and Diehl, 2004).

Due to the increasing use of ASs, they have been widely detected in wastewaters (Loos et al., 2013) and environmental matrices, including surface water, underground water, and precipitation and soil (Scheurer et al., 2009; Gan et al., 2013b; Gan et al., 2014a; Tran et al., 2014). Among ASs, SAC, SUC, cyclamate (CYC) and acesulfame (ACE) are typical ASs that have been widely detected. Due to their great water solubility (from 4 to 1000 g/L at 25 °C) (TOXNET, 2017), aquatic environment is the main sink of ASs, which have been widely reported (Scheurer et al., 2009; Buerge et al., 2011; Kokotou et al., 2012; Sang et al., 2014; Watanabe et al., 2016; Yin et al., 2017). ASs have been reported to occur at ng/L to mg/L levels in wastewaters (Loos et al., 2013) and landfill leachates (Roy et al., 2014), and at ng/L to µg/L levels in river and coastal waters (Mead et al., 2009; Gan et al., 2013b), drinking water (Scheurer et al., 2010; Mawhinney et al., 2011), rainwater (Oppenheimer et al., 2012), and groundwater (Gan et al., 2013b; Tran et al., 2014). Though their water solubility is high, the ASs were also detected in sewage sludge (Ordonez et al., 2013; Subedi et al., 2014a) and suspended particulate matter (SPM) of wastewater (Subedi and Kannan, 2014b). Though very limited, several papers studied the toxicity of ASs on ecosystem. SUC in the aquatic environment have been showed to alter the physiology and locomotion of *Daphnia magna* (Huggett and Stoddard, 2011; Wiklund et al., 2012). SUC were also reported to be able to interfere with photosynthesis in algae (Lubick, 2008). However, the effective concentrations in these studies are mg/L levels, much higher than the typical concentrations of ASs in the environment. Further studies should be conducted to reveal the possible long term effects of AS


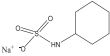

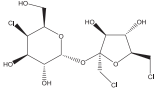
on specific aquatic organisms at environmentally relevant concentrations (Lange et al., 2012; Stolte et al., 2013).

Studies on the environmental fate of ASs have revealed that ACE and SUC are persistent and hardly eliminated in municipal wastewater treatment plants (WWTPs), with removal efficiencies below 20% (Lunden et al., 2008; Buerge et al., 2009; Scheurer et al., 2009; Gan et al., 2013b), whereas CYC and SAC were easily biodegraded but still detected in effluents as well as in other aquatic environments due to their huge loadings. It was found that the half-life of ACE in an urban river was 9 d under the joint effects of microorganisms and photodegradation, which was enhanced by dissolved organic matter (DOM) (Gan et al., 2014b). Due to their great solubility, the ASs are hardly bioaccumulated by aquatic organisms, e. g., bioconcentration factor (BCF) of SUC was calculated to be <1 for algae and between 1.6 and 2.2 for daphnids under two exposure concentrations (10 and 100 mg/L) (Lillicrap et al., 2011).

Dissimilar to the large numbers of literature on aquatic environments, thus far, only three studies have reported the occurrence of ASs in soil (Buerge et al., 2011; Gan et al., 2014a; Foolad et al., 2015). Various sources were proposed for the ASs in soil, including domestic or industrial sewage irrigation, pig manure or sludge fertilization, and metabolites of certain sulfonylurea herbicides (Roberts and Hutson, 1998; Buerge et al., 2011; Lange et al., 2012). Furthermore, ASs may also end up in the terrestrial environment via dry and wet deposition. A nationwide investigation in China conducted by our research group revealed the wide occurrence of ASs in atmospheric dust deposition with concentrations up to 6450 ng/g (Gan et al., 2013b); Moreover, up to 1.3 µg/L ASs were determined in rainfall in Tianjin, China in another study (Gan et al., 2014a). Pig farm is an important potential source for ASs in the environment due to their wide use as feed additives to improve the taste of pig feed. However, no data can be found in the literature on ASs' fate in a pig farm and neighboring soil system after the wastewater and manure fertilizers are applied to soil.

As for the fate of ASs in soil, only one group studied the dissipation of ASs in soil using laboratory batch incubation experiment and found that SAC, ACE, CYC, and SUC were degraded with half-lives of 3–12, 3–49, 0.4–6, and 8–124 days, respectively (Buerge et al., 2011). However, no study has examined the degradation of ASs in a field environment. The contamination of ASs in soil may pose a risk to groundwater due to their high water solubility. Indeed, SAC was detected in groundwater from a pumping station in Switzerland at concentrations up to 0.26 µg/L (Buerge et al., 2011). Four typical ASs, ranging from 7.1 to 100 ng/L, were determined in groundwater samples collected around Tianjin by our research group (Gan et al., 2014a). Recently, the bioaccumulation of emerging chemicals in plants has drawn much attention since these compounds are more likely to be absorbed by plants due to their high water solubility as compared to the traditional hydrophilic organic

Table 1
Structures and selected properties of the four typical artificial sweeteners detected widely in the environment.

	Saccharin	Cyclamate	Acesulfame	Sucralose
CAS no.	81-07-2	139-05-9	33665-90-6	56038-13-2
Structure				
Molecular weight (g/mol)	183.2	179.2(202.2)	163.2	397.6
Sugar equivalence	300	30	200	600
Water solubility (25 °C) (g/L)	4	1000	270	283
pK _a ^a	2.2	1.9	2	11.8
Log K _{ow}	0.91	-1.61	-1.33	-1
ADI ^b mg/kg body weight	5 (sodium salt), 3.8 (free acid)	7	9 (potassium salt)	15

Data were from TOXNET (<http://toxnet.nlm.nih.gov>);

ADI: acceptable daily intake.

^a data were cited from a previous study (Lange et al., 2012).

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