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Suitability of artificial sweeteners as indicators of raw wastewater contamination in surface water and groundwater

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ABSTRACT

There is no quantitative data on the occurrence of artificial sweeteners in the aquatic environment in Southeast Asian countries, particularly no information on their suitability as indicators of raw wastewater contamination on surface water and groundwater. This study provided the first quantitative information on the occurrence of artificial sweeteners in raw wastewater, surface water and groundwater in the urban catchment area in Singapore. Acesulfame, cyclamate, saccharin, and sucralose were ubiquitous in raw wastewater samples at concentrations in the range of ng/L–μg/L, while other sweeteners were not found or found only in a few of the raw wastewater samples. Residential and commercial effluents were demonstrated to be the two main sources of artificial sweeteners entering the municipal sewer systems. Relatively higher concentrations of the detected sweeteners were frequently found in surface waters at the sampling sites located in the residential/commercial areas. No significant difference in the concentrations of the detected sweeteners in surface water or groundwater was noted between wet and dry weather conditions (unpaired T-test, $p > 0.05$). Relatively higher concentrations and detection frequencies of acesulfame, cyclamate and saccharin in surface water samples were observed at the potentially impacted sampling sites, while these sweeteners were absent in most of the background surface water samples. Similarly, acesulfame, cyclamate, and saccharin were found in most groundwater samples at the monitoring well (GW6), which is located close to known leaking sewer segment; whereas these were absent in the background monitoring well, which is located in the catchment with no known wastewater sources. Taken together, the results suggest that acesulfame, cyclamate, and saccharin can be used as potential indicators of raw wastewater contamination in surface water and groundwater.

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1. Introduction

Environmental pollution by emerging organic contaminants such as pharmaceuticals and personal care products, and food additives has been increasingly gained attention due to their potential in causing undesirable ecosystem and human health effects (Daughton and Ternes, 1999; Huggett and Stoddard, 2011; Tran et al., 2013b). In recent years, it has been found the widespread presence of some artificial sweeteners such as acesulfame (ACE), saccharin (SAC), sucralose (SUC), aspartame (ASP), and cyclamate (CYC) in wastewater, groundwater, surface water, bank filtrate and drinking water systems (Buerge et al., 2009; Scheurer et al., 2009; Oppenheimer et al., 2011; Scheurer et al., 2011; Van Stempvoort et al., 2011; Lange et al., 2012). Artificial sweeteners end up in the environment through human consumption of low-calorie beverage, food and other products, in which sweeteners are used as sugar substitutes in remarkable amounts (Kroger et al., 2006). Consumed artificial sweeteners are excreted through urine or feces (Lange et al., 2012). The compounds are then carried to wastewater treatment plants (WWTPs) where some of them can be partially degraded or pass the process unchanged (Buerge et al., 2009; Scheurer et al., 2009, 2012). Previous studies found that several artificial sweeteners, such as SUC and ACE, were not or poorly degraded during wastewater treatment processes and were detected in municipal effluents, surface waters, and potable water at concentrations up to the μg/L level (Scheurer et al., 2009; Mawhinney et al., 2011; Oppenheimer et al., 2011; Gan et al., 2013). In a previous monitoring survey of groundwater near Zurich in Switzerland, Buerge et al. (2009) found the presence of ACE in 65% of the investigated groundwater samples and the highest concentration of SUC in groundwater, up to 4.7 μg/L, was observed in areas with significant infiltration of river water. In most studies in Switzerland, ACE was often present at higher level than SUC in the aquatic environment (e.g. wastewater, surface water and groundwater). In a survey of rivers in 27 European countries, Loos et al. (2009) examined 120 samples collected from the rivers and found that the concentration of SUC was predominantly observed in samples collected from the United Kingdom, Belgium, The Netherlands, France, Switzerland, Spain, Italy, Norway, and Sweden at concentration level up to 1 μg/L. While it was only detected at minor levels (<100 ng/L) in samples from Germany and Eastern Europe, implying a lower usage of SUC in those countries (Richardson and Ternes, 2011).

The first findings of the presence of artificial sweeteners in the aquatic environment in the United States was reported by Mead et al. (2009) when carrying out a survey of the occurrence of SUC in coastal and marine waters. Among the widely used artificial sweeteners, ACE and SUC showed to be the most persistent in natural waters as well as in wastewater/drinking water treatment processes (Scheurer et al., 2009, 2012). In studies in Europe, ACE was the only artificial sweetener found in finished drinking water with concentrations up to several hundred ng/L. While a recent study conducted in the U.S has found the occurrence of SUC in all tested drinking water treatment plants (DWTPs), the highest concentration in different DWTPs ranged from 100 ng/L to 2900 ng/L (Mawhinney et al., 2011). The fate of the artificial sweeteners during wastewater and drinking treatment processes has been reported in several studies (Mawhinney et al., 2011; Scheurer et al., 2011, 2012).

Due to their recalcitrance to transformation and low adsorption capacity on soils, ACE and SUC have been viewed as ideal markers for the evaluation of the impact of wastewater contamination on receiving water bodies, and particularly in groundwater (Buerge et al., 2009; Oppenheimer et al., 2011; Scheurer et al., 2011; Van Stempvoort et al., 2011). Buerge et al. (2009) also found that the concentrations of ACE in groundwater samples collected from the monitoring wells were similar to those of wastewater effluents, while concentrations of SUC in groundwater were significantly lower. SAC and CYC were, however, not observed in any groundwater samples. These results suggested that ACE could be considered as a potential tracer for assessing the impact of wastewater contamination in groundwater (Buerge et al., 2009). In a recent study in the United States, Oppenheimer et al. (2011) found that among 85 trace organic compounds investigated in different water samples, SUC was the most suitable indicator compound used to evaluate the impact of wastewater contamination on receiving water bodies. Although numerous studies on the occurrence and fate of artificial sweeteners in a variety of environmental compartments (such as wastewater, surface water, groundwater, and agricultural soil), most of these studies have been conducted in the areas where surface water and groundwater are impacted by the discharge of treated wastewater effluents into surface water bodies (i.e. rivers, canals, and lakes), runoffs from agricultural field irrigated with treated wastewater, infiltration from sewage disposal sites and septic tanks (Buerge et al., 2009; Mead et al., 2009; Scheurer et al., 2009; Buerge et al., 2011; Mawhinney et al., 2011; Oppenheimer et al., 2011; Van Stempvoort et al., 2011; Berset and Ochsenbein, 2012). As a result, artificial sweeteners were detected in surface waters and groundwater in those areas. To the best knowledge of the authors, there has been no or little information in the open literature documenting the occurrence of artificial sweeteners in surface water and groundwater in highly urbanized areas like Singapore where both surface water and groundwater are not impacted by the discharge of treated wastewater effluents since there are separate systems to collect stormwater and raw wastewater in Singapore, and treated wastewater effluents from wastewater treatment plants (WWTPs) are directly discharged into the sea. Additionally, both surface water and groundwater in Singapore are not much impacted by runoff from agricultural fields irrigated with treated wastewater, infiltration of sewage disposal sites or riverbank filtration. Thus, the distribution of artificial sweeteners in surface water and groundwater may be different from those countries discussed above. The first objective of this study aims to fill the existing gap by providing the first and comprehensive data on the occurrence of the six widely used artificial sweeteners in raw wastewater, surface waters, and groundwater in Singapore where food and drinking cultures are different