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Degradation of sucralose in groundwater and implications for age dating contaminated groundwater

W.D. Robertson [a], *, D.R. Van Stempvoort [b], J. Spoelstra [a], [b], S.J. Brown [b], S.L. Schiff [a]

[a] University of Waterloo, Waterloo, Ontario, N2L 3G1, Canada
[b] Environment Canada, Burlington, Ontario, Canada

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ABSTRACT

The artificial sweetener sucralose has been in use in Canada and the US since about 2000 and in the EU since 2003, and is now ubiquitous in sanitary wastewater in many parts of the world. It persists during sewage treatment and in surface water environments and as such, has been suggested as a powerful tracer of wastewater. In this study, longer-term persistence of sucralose was examined in groundwater by undertaking a series of three sampling snapshots of a well constrained wastewater plume in Canada (Long Point septic system) over a 6-year period from 2008 to 2014. A shrinking sucralose plume in 2014, compared to earlier sampling, during this period when sucralose use was likely increasing, provides clear evidence of degradation. However, depletion of sucralose from a mean of 40 μg/L in the proximal plume zone, occurred at a relatively slow rate over a period of several months to several years. Furthermore, examination of septic tank effluent and impacted groundwater at six other sites in Canada, revealed that sucralose was present in all samples of septic tank effluent (6–98 μg/L, n = 32) and in all groundwater samples (0.7–77 μg/L, n = 64). Even though sucralose degradation is noted in the Long Point plume, its ubiquitous presence in the groundwater plumes at all seven sites implies a relatively slow rate of decay in many groundwater septic plume environments. Thus, sucralose has the potential to be used as an indicator of 'recent' wastewater contamination. The presence of sucralose identifies groundwater that was recharged after 2000 in Canada and the US and after 2003 in the EU and many Asian countries.

1. Introduction

Some trace organic constituents that are unique to sanitary wastewater, persist during sewage treatment and in aqueous environments and consequently, several are being used as tracers to fingerprint the presence of sanitary wastewater in surface water and groundwater environments. These tracers include the pharmaceutical compound carbamazepine (e.g. Clara et al., 2004; Van Stempvoort et al., 2013) and the artificial sweetener potassium acesulfame (e.g. Buerge et al., 2009; Spoelstra et al., 2013), widely used in diet soft drinks and other food products. These two compounds have garnered attention because they occur in sanitary wastewater at concentrations that are orders of magnitude above background levels and thus, they offer the potential to reveal the presence of even very small amounts of wastewater. For example, in a screening survey of 100 wells in Switzerland, acesulfame (ACE) was detected in 65 of the samples (Buerge et al., 2009) and in a study of a urban groundwater in Canada, ACE was detected in >50% of the samples from seven different sites (Van Stempvoort et al., 2011a). Wolf et al. (2012) measured a suite of 71 trace organic constituents in a network of monitoring wells testing for sewer leakage in the city of Rastatt, Germany and detected carbamazepine and ACE most frequently, (33 and 28% of samples respectively).

Another commonly used artificial sweetener, sucralose (SUC), has also become ubiquitous in sanitary wastewater in many parts of the world. It too exhibits minimal removal during sewage treatment (Buerge et al., 2009; Schmid Neset et al., 2010; Lange et al., 2012; Tran et al., 2014; Subedi and Kannan, 2014) and during the production of potable water supplies (Mawhinney et al., 2011) and consequently, has also been proposed as a tracer for detecting sanitary wastewater. Oppenheimer et al. (2011) measured a suite of 85 trace organic and pharmaceutical compounds in a variety of US sanitary wastewaters and concluded that SUC was the best indicator parameter because it was detected in all effluent samples

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* Corresponding author. Dept. Earth and Environmental Sciences, University of Waterloo, Waterloo, ON, N2L 3G1, Canada.
E-mail address: wroberts@uwaterloo.ca (W.D. Robertson).

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(mean of 27 μg/L, n = 16) and in all surface water samples downstream of wastewater treatment plants (0.12–10 μg/L, n = 11), whereas it was not detected in surface waters where wastewater input was absent (<0.1 μg/L, n = 15). All other compounds, including carbamazepine, were detected less frequently in the wastewater-impacted streams and many were prone to false positives in the non-impacted streams. Acesulfame was not assessed in the above study, however.

Another consideration for SUC is that it is the newest of the commonly used artificial sweeteners, having only been approved for use in the US in 1998 and in the EU in 2003 (Tate and Lyle PLC, 2014). Large scale commercial production of SUC began in 2000 (Tate and Lyle PLC, 2014) and by 2006, it was the largest selling artificial sweetener in the US (Browning, 2007). Thus, over the last decade or so, SUC concentrations in sanitary wastewaters have likely increased considerably. If SUC is as equally mobile and persistent in soils as is ACE, as some laboratory studies suggest (Buerge et al., 2011), then recent increases in SUC concentrations in wastewater could provide a means of providing an approximate age date for wastewater contaminated groundwater, an often problematic task. For example, if a well sample showed evidence of wastewater because of the presence of artificial sweeteners, if the ratio of SUC/ACE was found to be similar to current wastewaters, then the contaminant source may only be a few years old. If the ratio was lower or if SUC was absent, this could indicate older impact originating prior to 2000 in the US or 2003 in the EU, and consequently such a well might be considered less at risk from wastewater-derived contaminants such as pathogens.

In our previous examination of a suite of four artificial sweeteners (acesulfame, sucralose, cyclamate and saccharin) in the Long Point septic system plume, ACE was found to be the most persistent, occurring throughout the 200 m mapped length of the plume (Robertson et al., 2013). Sucralose was also present in concentrations similar to ACE (30–80 μg/L) in the plume proximal zone, but concentrations declined abruptly to close to the method detection limit (5 μg/L at that time), about 70 m from the tile bed in a plume zone that was about 5 yrs old. This zone would have originated around 2005 (sampled in 2010), which is the time frame when SUC use was likely increasing rapidly in Canada (Supporting Material). Although it was previously suggested that the diminished size of the SUC plume at Long Point was evidence of degradation or adsorption (Van Stempvoort et al., 201lb; Robertson et al., 2013), lower historical usage of SUC could have also produced the same effect. Batch tests conducted by LaBare and Alexander (1993) did show degradation of SUC in organic carbon-rich soil and lake bottom sediments (1–17 wt% OC), with 50% removal occurring in about 30–100 days. Much less removal was noted however, in lower carbon, lake water samples (<4% over 80 days, DOC = <50–62 mg/L), and rates were reduced or negligible under anaerobic conditions (LaBare and Alexander, 1993). During these tests, SUC loss was noted immediately, with no lag period, indicating that it was likely cometabolized with other carbon sources.

For the current study, the Long Point septic system plume was resampled and analysed for artificial sweeteners in 2014. This provided a third detailed sampling snapshot over this critical period from 2008 to 2014, shortly after when SUC use was increasing. Examination of concentration trends over this time period should provide insight as to whether the SUC plume at Long Point is dominated by degradation or alternatively, by changing usage patterns. In addition, we analysed septic tank effluent and associated septic system-impacted groundwater, at six other sites in Ontario, Canada, to establish a range of SUC concentrations in typical sanitary wastewater and to examine its persistence in a variety of subsurface environments.

2. Study sites

2.1. Long Point site

The Long Point (LP) campground, located on the north shore of Lake Erie, has 256 overnight campsites and is open seasonally from mid-May until mid-October. Sewage from a single washroom facility is treated on-site, in a conventional septic system consisting of a septic tank and two separate infiltration beds (tile beds), ~290 m^2 each, that receive average wastewater loading of 6 cm/d during the peak use months of July and August. Water limiting fixtures in the washroom result in relatively high concentrations of constituents such as NH_4^+-N (100 ± 27 mg/L) and ACE (50 ± 15 μg/L, Robertson et al., 2013) in the wastewater. The groundwater plume from Tile Bed 2 flows southward toward the Lake Erie shoreline within a ~5 m thick unconfined calcareous sand aquifer that is underlain by a clayey silt unit (Fig. 1). In a previous study (Robertson et al., 2013) it was determined that Cl^-, Na^+ and ACE could be used to map the wastewater plume, although high background Cl^- values in down gradient areas obscured the Cl^- plume there. Except within about 0.5 m of the water table, groundwater throughout the site, both in and outside of the wastewater plume, is sub-oxic (dissolved oxygen (DO) < 1 mg/L, Robertson et al., 2012). In a July 2008 tracer test, NaBr was injected into the septic tank and 290 days later, elevated Br was detected up to 15 m down-gradient from the tile bed (Robertson et al., 2012), indicating a horizontal groundwater velocity of 22 m/yr in the tile bed area. The 200 m-long monitored section of the plume (Fig. 1) is about 15 years old based on tritium/helium age dating undertaken in 2011 (Robertson et al., 2013). Groundwater monitoring in the Tile Bed 2 area has been ongoing since the septic system was commissioned in 1991 (e.g. Aravena and Robertson, 1998; Van Stempvoort et al., 2011b).

2.2. Other sites

The other six septic system sites examined in this study are also located Ontario and include two large seasonal-use campgrounds (KP and KR sites), a seasonal-use tourist resort (LJ site), a communal townhouse complex (BU site) and two seasonal-use family cottages (JL and GB sites). All of these septic systems are at least 10 years old and are generally of conventional design, consisting of a septic tank and associated infiltration bed with perforated infiltration pipes installed into the vadose zone at 0.5–1 m depth below ground surface. Two of the sites (family cottage, GB and communal residence, BU) also provide tertiary treatment using aerobic fixed bed filters prior to discharge to the infiltration beds, and the latter site also includes a denitrification module. Most of these are communal systems that receive moderate wastewater loading rates in the range 0.4–6 cm/d during peak use in the summer (July and August). The two family cottage sites (GB and JL) receive slightly lower loading of ~0.3 cm/d during July and August. The effluent at these sites can be considered typical of sanitary wastewater generated in Canada, except that most of these, excluding the townhouse site (BU), are seasonal-use systems that receive highest loading during the summer months. This could influence artificial sweetener concentrations, as there is likely increased consumption of certain food products such as diet soft drinks during the summer. Additional site details are provided in the Supporting Material.

3. Methods

3.1. Sampling and analyses

In the current study, most sampling occurred from June, 2011–January, 2014, but earlier results from the Long Point site