GROUNDWATER QUALITY

Artificial Sweeteners Reveal Septic System Effluent in Rural Groundwater

John Spoelstra,* Natalie D. Senger, and Sherry L. Schiff

Abstract

It has been widely documented that municipal wastewater treatment plant effluents are a major source of artificial sweeteners to surface waters. However, in rural areas, the extent to which septic systems contribute these same compounds to groundwater aguifers is largely unknown. We examined the occurrence of four commonly used artificial sweeteners in an unconfined sand aquifer that serves as a water supply for rural residents, as a receptor of domestic wastewater from septic systems, and as a source of baseflow to the Nottawasaga River, ON, Canada. Groundwater from the Lake Algonquin Sand Aquifer in the southern Nottawasaga River Watershed was collected from private domestic wells and as groundwater seeps discharging along the banks of the Nottawasaga River. Approximately 30% of samples had detectable levels of one or more artificial sweeteners, indicating the presence of water derived from septic system effluent. Using acesulfame concentrations to estimate the fraction of septic effluent in groundwater samples, ~3.4 to 13.6% of the domestic wells had 1% or more of their well water being derived from septic system effluent. Similarly, 2.0 to 4.7% of the groundwater seeps had a septic effluent contribution of 1% or more. No relationship was found between the concentration of acesulfame and the concentration of nitrate, ammonium, or soluble reactive phosphorus in the groundwater, indicating that septic effluent is not the dominant source of nutrients in the aquifer. It is expected that the occurrence of artificial sweeteners in shallow groundwater is widespread throughout rural areas in Canada.

Core Ideas

• Samples were collected from domestic wells and groundwater seeps in rural Ontario.

• Artificial sweeteners were found in >30% of rural groundwater samples.

• Acesulfame was used to estimate the fraction of septic effluent in groundwater.

• Septic effluent was not a significant source of nutrients in the aquifer.

Copyright © American Society of Agronomy, Crop Science Society of America, and Soil Science Society of America. 5585 Guilford Rd., Madison, WI 53711 USA. All rights reserved.

J. Environ. Qual. 46:1434–1443 (2017). doi:10.2134/jeq2017.06.0233 This is an open access article distributed under the terms of the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/). Received 9 June 2017. Accepted 3 Oct. 2017. *Corresponding author (John.Spoelstra@canada.ca). N RURAL AREAS, municipal water supplies and wastewater treatment facilities are generally not available, and therefore rural households rely on private wells for water supply and treat their wastewater using septic systems. By design, septic systems discharge treated wastewater to groundwater. Groundwater contamination by septic effluents and associated constituents (e.g., nitrate, personal care products, pharmaceuticals, pathogenic microorganisms) has been recognized as a potential health concern in rural areas (Withers et al., 2014). In the case of surficial aquifers, the separation between wastewater discharge areas and the intakes for water supply wells can be minimal, increasing the chances of wells intercepting groundwater contaminated by septic effluent.

In Canada and the United States, 11 (Statistics Canada, 2011) and 14% (Maupin et al., 2014) of households, respectively, get their water from private wells. Private well owners are not subject to the same regulations and monitoring that municipalities are with respect to the quality of their water supplies. For example, the maximum acceptable concentration for nitrate in Canada is 10 mg N L⁻¹. Despite this, many studies have shown that groundwater used for private water supplies in rural areas often has nitrate concentrations that exceed the drinking water limit (Hill, 1982; Goss et al., 1998). Municipal water supplies are often sourced from groundwater and are therefore also not immune to impact from septic system effluents. As urban areas expand into formerly rural areas, previous land use activities such as agriculture and septic systems are likely to influence the quality of groundwater resources in areas where new municipal wells are located.

Studies have used a variety of methods to investigate potential septic system impacts to groundwater and to estimate the fraction of private wells affected by septic effluent. One such method is to analyze well samples for total coliform bacteria, which include fecal coliforms that reside in the digestive tracts of humans and animals and are also found in septic effluent. The presence of living coliforms in water indicates relatively recent contamination of the water and the possible presence of other bacteria and viruses that could have human health consequences if ingested. For example, Francy et al. (2000) found that 20% of

J. Spoelstra, Environment and Climate Change Canada, Canada Centre For Inland Waters, 867 Lakeshore Rd., PO Box 5050, Burlington, ON, Canada L75 1A1; J. Spoelstra, N.D. Senger, and S.L. Schiff, Dep. of Earth and Environmental Sciences, Univ. of Waterloo, 200 University Ave. W, Waterloo, ON, Canada N2L 3G1. Assigned to Associate Editor Clinton Williams.

Abbreviations: ACE, acesulfame; CYC, cyclamate; IC, ion chromatography; LASA, Lake Algonquin Sand Aquifer; mdl, method detection limit; MS/MS, tandem mass spectrometry; OWCs, organic wastewater compounds; SAC, saccharin; SRP, soluble reactive phosphorus; SUC, sucralose.

143 groundwater samples collected in the United States as part of their study tested positive for total coliforms. In a study of 1292 domestic wells on Ontario farms, Goss et al. (1998) found that 34% of the wells tested had coliform bacteria counts in excess of the maximum acceptable concentration.

Since fecal coliforms are present in both human and animal waste, their presence in groundwater is not strictly indicative of septic-derived contamination. In rural areas where septic systems and livestock operations are often both located, other methods may be required to attribute fecal contamination to a specific source. One such technology is microbial source tracking, which can be used to trace the origin of fecal bacteria and viruses to specific animals (e.g., humans, cows, pigs, ducks, etc.; Khan et al., 2007; Roslev and Bukh, 2011; Rusiñol et al., 2014).

The presence of certain pharmaceutical compounds in groundwater is also used to detect the presence of humanderived wastewater. Pharmaceuticals that are often detected with sufficient frequency and concentration to be considered useful tracers of wastewater-affected groundwater include carbamazepine, primidone, sulfamethoxazole, cotinine, acetaminophen, and caffeine (Seiler et al., 1999; Drewes et al., 2002; Barnes et al., 2008; Fram and Belitz, 2011; Van Stempvoort et al., 2013; Schaider et al., 2014).

Numerous studies have now demonstrated that artificial sweeteners are powerful tracers of wastewater in the environment, partly due to their widespread inclusion in products such as diet beverages and foods, pharmaceuticals, and toothpaste. Artificial sweeteners have several advantages over other wastewater tracers, including the facts that (i) they are almost exclusively derived from human wastewater (municipal or septic system effluents), (ii) they are ubiquitous in human wastewater, (iii) they occur at relatively high concentrations, and (iv) they are slow to breakdown in the environment, especially acesulfame (ACE) and sucralose (SUC). Artificial sweeteners have been detected in rivers, lakes, and groundwater where wastewater is known to discharge (Brorström-Lundén et al., 2008; Buerge et al., 2009; Scheurer et al., 2009; Oppenheimer et al., 2011; Spoelstra et al., 2013; Robertson et al., 2013; Van Stempvoort et al., 2013). They have also been found in the groundwater underlying urban areas with leaky sewer systems (Wolf et al., 2012; Tran et al., 2014; Lee et al., 2015) and as a component of landfill leachate (Roy et al., 2014; Clarke et al., 2015).

Once in the environment, numerous factors can control the fate of artificial sweeteners. Saccharin (SAC) and cyclamate (CYC) have generally been found to be more easily degraded than SUC and ACE (Buerge et al., 2009; Scheurer et al., 2009; Robertson et al., 2013) and have been proposed as tracers of untreated or undertreated wastewater (Buerge et al., 2009; Yang et al., 2017). For example, Zirlewagen et al. (2016) used CYC/ACE ratios to distinguish between "old or treated wastewater" and "fresh and untreated wastewater." The presence of the more degradable artificial sweeteners in groundwater can be an indication of more recent contamination and proximity to the effluent source.

Although previous studies documented minimal ACE degradation in wastewater treatment plants, (Buerge et al., 2009; Scheurer et al., 2009), recent studies (Castronovo et al., 2017; Yang et al., 2017) found that ACE degradation can be significant in some wastewater treatment systems. Even so, Yang et al. (2017) show that ACE concentrations in effluent can remain very high compared with other potential organic wastewater tracers, only being surpassed by erythromycin-H₂O and SUC in a study of nine wastewater treatment plants in China. Acesulfame appears to be very resistant to degradation once released into the environment and therefore can be used to track wastewater far from its source. For example, Spoelstra et al. (2013) demonstrated that, after discharge from wastewater treatment plants, ACE can persist for hundreds of kilometers in rivers. In groundwaters, previous studies have shown that ACE is more persistent than SUC (Scheurer et al., 2009; Robertson et al., 2013). Acesulfame also survives water treatment processes and can subsequently end up in the municipal water supply for communities sourcing their raw municipal water from surface waters and groundwaters containing wastewater (Buerge et al., 2009; Scheurer et al., 2010; Prasse et al., 2011; Gan et al., 2013; Nödler et al., 2013; Spoelstra et al., 2013; Ens et al., 2014; Scheurer et al., 2014). Aspartame, another commonly used artificial sweetener, is metabolized completely within the body (Renwick, 1986) and is therefore not found in wastewater-affected environmental samples (Scheurer et al., 2009; Lange et al., 2012).

We hypothesize that a significant fraction of shallow groundwater in unconfined aquifers in rural settings, where septic systems are common, is affected by septic system effluent and that analysis of artificial sweeteners in groundwater will indicate this. However, in these areas where intensive agriculture activities also take place, we suspect that septic effluents are not a significant source of nitrate and other nutrients in regional groundwater, in comparison with that derived from agriculture (e.g., fertilizer application to crops). This study examines the occurrence and concentration of four commonly used artificial sweeteners (ACE, SAC, CYC, SUC) in a surficial aquifer in a rural setting that functions both as a source of water for private domestic wells and also receives treated wastewater effluent from septic systems. Furthermore, for groundwater wells and seeps containing artificial sweeteners, we use the concentration of ACE to estimate the fraction of water derived from septic effluents and to see if nutrients in groundwater are related to the degree of septic influence.

Materials and Methods Site

The occurrence of artificial sweeteners in rural groundwater was investigated in the Nottawasaga River Watershed, located ~50 km north of Toronto, ON, Canada. There are more than 10,000 privately owned wells in the Nottawasaga River Watershed and greater than 130 municipal wells, which provide domestic water to most of the watershed's residents (Nottawasaga Valley Conservation Authority, 2013). The Nottawasaga River has its headwaters in the Oak Ridges Moraine and discharges to Georgian Bay at Wasaga Beach. Land use in the Nottawasaga River Watershed (3147 km²) consists largely of agriculture (56%), with potatoes (Solanum tuberosum L.) and sod being major crops in the southern portion of the watershed (Van Stempvoort et al., 2016) where groundwater was collected for this study. The basin has a temperate climate and receives ~790 mm of precipitation annually, with ~20% coming in the form of snow (Environment and Climate Change Canada, 2017). The surficial geology of the southern Nottawasaga River Watershed is dominated by the Lake Algonquin Sand Aquifer (LASA; Sibul and Choo-Ying,

1971), which is an unconfined sand aquifer that supplies water for domestic and agricultural use in the rural areas. The thickness of the LASA varies but is generally ~12 m, with a maximum thickness ~25 m within our study area (Sibul and Choo-Ying, 1971). The contact between the LASA and underlying finergrained strata is exposed along the incised banks of the southern Nottawasaga River. As such, groundwater discharge to this section of the river occurs above the river water level and subsequently runs down the banks, facilitating groundwater sampling along the seepage faces (Hill, 1982).

Sampling

Groundwater samples from the southern Nottawasaga River Watershed were collected from seeps discharging along the banks of the Nottawasaga River and from private domestic wells (Fig. 1). Using a small boat, groundwater seeps were spotted along the banks of the river and bulk samples collected and kept in coolers with ice packs until further processing (e.g., subsampling, filtering, preserving) later that day (within 12 h). Groundwater seeps were collected along the southern section of the Nottawasaga River between Tottenham Road ($44^{\circ}07'35''$ N, $79^{\circ}50'03''$ W) and Murphy Road ($44^{\circ}14'58''$ N, $79^{\circ}49'18''$ W) in September 2010 and May 2011 and between Line 5 ($44^{\circ}10'39''$ N, $79^{\circ}49'01''$ W) and Murphy Road in June 2013. Groundwater from private wells was collected in June and July 2011 from taps that, where possible, bypassed any treatment systems (e.g., water softeners). Field parameters (temperature, pH, conductivity, dissolved oxygen) were monitored with a YSI 600QS Sonde while the taps were allowed to run for several minutes until these parameters stabilized. Once stable, a bulk water sample was collected and then immediately subsampled for various chemical and stable isotopic parameters (not all reported here; Senger, 2016). Samples were kept cold until analysis, either refrigerated (4°C) or frozen (-18° C), depending on the parameter (see below).

Chemical Analyses

Major anion samples were filtered to $0.45 \,\mu m$ and refrigerated until analysis (within 1 wk) using a Dionex 2500 ion chromatograph. Sample concentrations were calibrated against certified multi-ion standards that were analyzed with the samples. The method detection limit (mdl) for nitrate using this technique was $0.016 \text{ mg N L}^{-1}$.

Subsamples for soluble reactive phosphorus (SRP) were field filtered to 0.45 μ m and refrigerated until analysis (within 48 h). The SRP concentrations were determined using an orthophosphate colorimetric technique, measuring the absorbance at 885 nm using a 10-cm, path-length cuvette on a Thermo



Fig. 1. Map of the study area. The green shaded area encompasses the area where domestic wells were sampled. Groundwater seep samples were collected along the highlighted (purple) portion of the Nottawasaga River. The inset map shows the location of Nottawasaga River Watershed within southern Ontario, Canada.

Scientific Evolution 201 ultraviolet-visible spectrophotometer. Sample values were calibrated against multiple SRP standards that were analyzed with the samples.

Ammonium subsamples were filtered to 0.45 μ m and acidified with hydrochloric acid to a pH of ~5 to 6 and stored frozen until analysis (within 48 h). Ammonium concentrations were determined using a colorimetric method (salicylate-nitroprusside) by measuring absorbance at 640 nm on a Beckman-Coulter DU720 ultraviolet-visible spectrophotometer. Sample values were calibrated against multiple ammonium standards that were analyzed with the samples.

The concentrations of four artificial sweeteners, ACE, CYC, SAC, and SUC, were determined using the methods described in detail by Van Stempvoort et al. (2011b, see supplemental information therein). In brief, 100 µL of sample was analyzed using suppressed ion chromatography (IC) coupled to a tandem mass spectrometer (MS/MS). Separation was performed using a Thermo Fisher Dionex 2500 IC system on a Dionex IONPAC AS20 analytical column (2×250 mm). The IC was interfaced to an AB Sciex 5500 QTrap MS/MS and operated in the negative electrospray ionization mode. Isotopically labeled compounds were used as internal standards to account for matrix effects. The domestic well samples (June and July 2011) and the groundwater seep samples collected in September 2010 and May 2011 were analyzed using the above technique, which had method detection limits of: ACE = 0.008, SAC = 0.021, CYC = 0.003, SUC = 5.0 μ g L⁻¹. The groundwater seep samples collected in September 2013 were run with a slightly modified method that included separation using a Thermo Fisher Dionex ICS 5000 system and ion suppression using a Dionex AERS 500 2 mm instead of a Dionex ASRS 300 2 mm. As a result of the method improvements, method detection limits were lowered for all but CYC, and especially for sucralose (ACE = 0.002, SAC = 0.002, CYC = 0.003, $SUC = 0.020 \ \mu g \ L^{-1}$).

When necessary for any of the parameters listed above, samples were diluted with Milli-Q water to bring their concentration within the working range of the standards.

Results and Discussion

Occurrence and Concentration of Artificial Sweeteners in Rural Groundwater

At least one of the four artificial sweeteners was found in 32% of the 149 groundwater seep samples collected along the banks of the Nottawasaga River. Acesulfame was the most commonly occurring artificial sweetener, with 21% of all seep samples collected having detectable (>mdl) ACE (Fig. 2A and 2B). Acesulfame also had the highest concentration of the four artificial sweeteners measured, with a maximum value of 1.7 μ g L^{-1} (Fig. 2B). The high prevalence and concentration of ACE is likely due to its common use as an artificial sweetener in Canada, its strong resistance to degradation in the environment (Buerge et al., 2009; Scheurer et al., 2010; Gan et al., 2014), and its mobility in groundwater (Buerge et al., 2009; Van Stempvoort et al., 2011a, 2011b; Robertson et al., 2013). Sucralose was analyzed for a smaller number of samples (only the 2013 set, n = 58), but it had the next highest overall occurrence and maximum concentration at 14% of samples and 0.57 µg L⁻¹, respectively (Fig. 2A and 2B). Saccharin and CYC were detected in 11 and





7% of samples, respectively, with concentrations as high as 0.095 and 0.18 μg $L^{-1},$ respectively (Fig. 2A and 2B).

The differences in method detection limits between the different sample sets had mixed consequences for the results. For ACE, only one seep sample from the 2013 set had a value between the old $(0.008 \,\mu\text{g L}^{-1})$ and new $(0.002 \,\mu\text{g L}^{-1})$ mdl, and therefore method improvements likely had a minimal effect on the frequency of ACE detections. In contrast, 14 of the 17 seep samples from 2013 that had detectable SAC were at concentrations that would not have been detected using the older method (i.e., $<0.021 \ \mu g \ L^{-1}$). Therefore, the change in mdl is likely the reason for the much higher percentage of SAC detections in the 2013 dataset (28%), compared with the 2010 and 2011 datasets (1%). The mdl for CYC was the same for both versions of the artificial sweetener analysis method. Considering only the seep data from 2013 (Fig. 2B), SAC actually had the highest occurrence of detections, but most were at very low concentrations. Considering only the 2013 results, 26% of the seep samples collected had one or more of the non-SAC artificial sweeteners detected, indicating some contribution of water derived from septic system effluent.

Similar to the groundwater seeps, at least one of three artificial sweeteners (SUC not reported) was found in 31% of the groundwater samples collected from 59 different private wells. Again, ACE had the highest occurrence and maximum concentration at 27% and 26.4 μ g L⁻¹, respectively (Fig. 2C). Saccharin and CYC detections were less frequent, at 5 and 8%, respectively, but maximum concentrations (SAC = 0.40, CYC = 4.1 μ g L⁻¹) were higher than those observed for groundwater seeps (Fig. 2C). In the groundwater seeps, SUC was only detected in samples that also had detectable ACE, and therefore we suspect that the percentage of domestic well samples having one or more artificial sweeteners was not affected by the exclusion of SUC. In contrast, the fraction of wells having artificial sweeteners might have been somewhat higher if analyzed using the newer (lower mdl) method, due to the likelihood of more SAC detections.

The nearly identical fraction of groundwater samples having measureable artificial sweeteners, whether from domestic wells or groundwater seeps discharging along the Nottawasaga River, suggests that these values may be representative of groundwater septic influence at the regional scale in the rural portions of the LASA and possibly other similar aquifers in Canada. To date, we are only aware of two previously published studies that have examined the occurrence of artificial sweeteners in groundwater from private domestic wells. Wu et al. (2014) sampled 100 domestic wells in Alberta, Canada, for ACE and SUC and found that 24 and 8% of the wells, respectively, had measureable ACE (9 imes 10⁻⁴ to 1.53 μ g L^{-1}) and SUC (0.065 to 0.54 µg L^{-1}). As with our groundwater seeps, Wu et al. (2014) found that SUC was only detected in samples that also contained ACE. Schaider et al. (2016) analyzed for a variety of organic wastewater compounds (OWCs), including ACE, in 20 domestic supply wells drawing water from a sand and gravel aquifer in Cape Cod, MA. Their sampling deliberately targeted wells with nitrate contamination, and therefore, as they acknowledge, their percentage of ACE detections is not representative of overall groundwater quality in the area. Of the 27 different OWCs detected, ACE was the most frequently detected (85% of wells sampled), had the highest concentration (5.3 μ g L⁻¹), and was always present if any other OWC was detected (Schaider et

al., 2016). Neither of the two abovementioned studies measured CYC or SAC concentrations.

Septic systems are designed to discharge effluent to the shallow groundwater, and given that artificial sweeteners, particularly ACE and SUC, are recalcitrant and mobile in groundwater, it is not surprising that artificial sweeteners are detected in aquifers receiving septic effluent. The LASA, being a highly permeable surficial aquifer, is vulnerable to surface-derived contaminants, including those from septic systems and land management practices (e.g., agriculture). For domestic wells for which total depth information was available (39 of 59 wells), ACE detections were limited to wells of 20-m depth below ground surface or less (Fig. 3). Of these 29 wells, 11 had detectable ACE concentrations. Groundwater that predates the widespread use of artificial sweeteners will not contain artificial sweeteners, even if once affected by septic system effluent. In Canada, SAC and CYC have been in use since at least the 1970s, whereas ACE and SUC were introduced later, in 1988 and 1992, respectively (Gougeon et al., 2004). The lack of ACE detections in wells known to be >20 m in the LASA could be due to the age of the groundwater at depth. Although there appears to be a general increase in ACE concentration with depth for those wells where ACE was detected (Fig. 3), interpretation of the concentration-depth relationship is complicated by the fact that we do not have information on the depth to the water table for each well, nor do we know the intake depths for the wells. For example, a well with a total depth of 20 m could be drawing groundwater from anywhere between the water table and 20 m if it is screened over that entire interval.

Although biosolids are occasionally applied in the Nottawasaga River Watershed, biosolids are not expected to be a significant source of artificial sweeteners to groundwater. Artificial sweeteners are highly soluble and therefore remain in the liquid phase of



Fig. 3. Concentration of acesulfame (ACE) with respect to the approximate domestic well depth (for wells where this information was available, 39 of 59 wells). The additional *y*-axis on the left shows the depth, in units of meters below ground surface (mbgs), for wells that did not have detectable ACE (i.e., below the method detection limit [mdl] of 0.008 μ g L⁻¹).

sewage, with relatively low amounts found in dewatered sewage sludge that biosolids are derived from (Pasquini et al., 2013; Subedi et al., 2014; Tran et al., 2014, 2015).

Piglet feed can contain SAC as an additive (Buerge et al., 2011), and therefore manure from swine farms can potentially be a source of SAC to the environment. However, it is unlikely that pig manure was a significant contributor of SAC to groundwater in our study area. First, there are very few pig farms in Simcoe County, where this study was conducted (Ontario Ministry of Agriculture Food and Rural Affairs 2016a, 2016b). Also, to our knowledge, the only artificial sweetener used in livestock feed is SAC, and therefore groundwater samples containing artificial sweeteners other than, or in addition to, SAC must be influenced by human wastewater. Saccharin is also produced from the breakdown of some sulfonylurea herbicides (Berger and Wolfe, 1996). Sulfonylurea herbicides are used in the Nottawasaga River Watershed, and therefore this is another potential source of SAC to groundwater in the area. Of the groundwater samples that contained artificial sweeteners, none of the domestic wells contained only SAC. Although eight of the groundwater seep samples contained only SAC, the concentrations were very low, ranging from 0.002 to $0.016 \,\mu g \, L^{-1}$.

The four artificial sweeteners measured in this study are all approved for human consumption by Health Canada, and therefore their presence in rural groundwater and private wells might not be considered a human health issue in and of itself. However, artificial sweeteners in groundwater indicate the presence of wastewater, which might be the source of other contaminants of concern (e.g., pathogens, nitrate, pharmaceuticals). For example, Zirlewagen et al. (2016) found a relationship between the occurrence of cyclamate and fecal indicator bacteria in a karst spring in southwestern Germany. Thus, artificial sweeteners can serve as a screening tool to identify groundwater that should receive additional testing, especially if the water is used for potable supplies. From an aquatic ecosystem perspective, the effects of environmentally relevant concentrations of artificial sweeteners in rivers and lakes are unknown for most organisms. However, once again, the presence of artificial sweeteners indicates a wastewater influence, and therefore, even if artificial sweeteners themselves are not a significant threat to the aquatic fauna, other compounds that are associated with wastewater might pose a threat and thus warrant further investigation. Furthermore, there is evidence that the breakdown products of artificial sweeteners might have ecotoxilogical implications (Sang et al., 2014; Li et al., 2016).

Fraction of Septic Effluent in Groundwater

The high occurrence of artificial sweeteners in rural groundwater in the southern Nottawasaga River Watershed demonstrates that a significant fraction of the rural groundwater is influenced by septic wastewater to some degree. Using ACE concentrations previously measured in septic tank effluents allows us to estimate the septic-derived water contribution in groundwater samples. In a study conducted in Ontario, the mean concentration of ACE in septic system effluent has been shown to range from 14 to 71 µg L⁻¹ (Robertson et al., 2016a, supporting information). Using these values as range limits, we calculated that 2.0 to 4.7% of the seeps and 3.4 to 13.6% of the domestic wells have 1% or more of their water being derived from septic system effluent (Fig. 4). Furthermore, the domestic wells with the two highest ACE concentrations of 5.4 and 26.4 μ g L⁻¹ have estimated septic water contributions of up to 38.3 and 100%, respectively (Fig. 4). Neither of these wells was being used for domestic drinking water supply at the time of sampling. The presence of artificial sweeteners in domestic water derived from groundwater dramatically illustrates a potential consequence of using the same aquifer for both wastewater treatment and water supply. Households with artificial sweeteners detected in their wells may be contaminating their water with their own septic effluent, or it might be derived from other households upgradient (i.e., their neighbors, as illustrated in the graphical abstract of Schaider et al., 2016).

The calculated septic water contributions shown in Fig. 4 are based on a couple key underlying assumptions. First, we assume that ACE concentrations in groundwater are not significantly affected by degradation. Currently, there are no studies that determine in situ ACE degradation rates in groundwater. However, ACE has been shown to be resistant to degradation in water and wastewater treatment plants, surface waters, and groundwater (Buerge et al., 2009; Scheurer et al., 2009; Robertson et al., 2013) and is subsequently very persistent in the environment. Acesulfame can be broken down by dissolved ozone (Scheurer et al., 2010, 2012), ultraviolet light (Sang et al., 2014; Scheurer et al., 2014; Li et al., 2016), and natural sunlight (Gan et al., 2014); however, these mechanism are not relevant to groundwater flow systems. A study using columns of saturated sandy soil has also shown that adsorption of artificial sweeteners (ACE, CYC, SAC) in sandy aquifers is likely negligible (Foolad et al., 2015). It should be noted that our septic water contributions represent minimum values, and if ACE degradation does occur in groundwater, then the actual septic water contributions would be higher than our calculated values.

Second, we assume that the septic tank ACE concentrations measured by Robertson et al. (2016a) are representative of septic system effluents in our study area. The availability of more septic effluent artificial sweetener concentrations will make these contribution calculations more robust in the future. At our current mdls, and the ACE concentrations found in septic effluent by Robertson et al. (2016a), our analytical method can detect a septic water contribution as low as 0.003% or ~1 mL $34 L^{-1}$ of water.

Nutrients from Septic System Effluent

For the southern Nottawasaga River, groundwater contribution to stream flow is obvious, as groundwater discharges through visible seeps along the river banks. Groundwater also contributes to streams and shorelines by discharging through the sediments below the waterline. Through its contribution along shorelines and as baseflow to streams, groundwater quality can influence surface water quality. Nitrate contamination of groundwater is known to be prevalent in the study area (Hill, 1982; Senger 2016), and although previously attributed to high fertilizer use by agriculture in the area, particularly potato farming, septic systems are another possible source of nitrate and other nutrients to groundwater.

As potential evidence for septic-derived nutrients, the relationships between groundwater nitrate, SRP, and $\rm NH_4^+$ concentrations and ACE were examined. Nitrate concentrations ranged from <mdl to 73 mg N L⁻¹ in the groundwater seeps and



Fig. 4. Estimated proportion of septic effluent in groundwater seeps (n = 149) and domestic wells (n = 59). The (A) minimum and (B) maximum effluent scenarios are based on mean septic effluent acesulfame concentrations of 71 and 14 µg L⁻¹, respectively (mean values for seven septic tanks, n = 37 in total; Robertson et al., 2016a).

from <mdl to 57 mg N L⁻¹ for domestic wells (Fig. 5A). Nitrate concentrations were not correlated with ACE concentration for seeps ($R^2 = 0.08$) or domestic wells ($R^2 = 0.15$). The full range of nitrate concentrations were observed for both seep and well samples in which ACE was not detected (i.e., <mdl) (Fig. 5A). With the exception of one seep sample (SRP = $134 \ \mu g \ L^{-1}$), the same was found for SRP (Fig. 5B). That is, the full range of concentrations was observed in samples in which ACE, and therefore septic influence, was not detected. For samples that did contain ACE, there was no correlation between the concentration of ACE and SRP ($R^2 = 0.02$ and 0.01 for seeps and wells, respectively; Fig. 5B). Groundwater ammonium concentrations were generally low for the seep samples, with 89% of samples having 0.1 mg NH₄⁺-N L⁻¹ or less, even if ACE was detected (Fig. 5C). The domestic well groundwater was also generally low in NH₄⁺, with only 17 samples having >0.1 mg NH₄⁺–N L^{-1} , and only four of these samples also having ACE (Fig. 5C). For samples that did contain ACE, there was no correlation between the concentration of ACE and NH_4^+ ($R^2 = 0.007$ and 0.002 for seeps and wells, respectively; Fig. 5C).

Extensive research of a groundwater septic plume at the Long Point Provincial Park in Ontario has shown that artificial sweeteners outlast many other septic system contaminants including nutrients (ammonium, nitrate, phosphorus) and pharmaceuticals (e.g., ibuprofen, naproxen, gemfibrozil) (Carrara et al., 2008; Robertson et al., 2013, 2016b). Therefore it is not surprising that we do not necessarily see a relationship between septic system nutrients and ACE in rural groundwater samples. The Long Point study site demonstrates the ability of artificial sweeteners to track septic system effluent water long after other wastewater constituents have disappeared, a characteristic that makes artificial sweeteners excellent tracers of wastewater in the environment and groundwater, specifically.

The lack of correlation between nitrate, ammonium, or SRP and the presence of ACE indicates that wastewater is not the dominant source of these compounds in groundwater at the



Fig. 5. Relationship between several potential septic-derived nutrient parameters: (A) nitrate, (B) soluble reactive phosphorus (SRP), and (C) ammonium and acesulfame (ACE) concentrations. The additional *y*-axes on the left show the parameter concentrations for samples that did not have detectable ACE (i.e., less than the method detection limit [mdl] of 0.008 μ g L⁻¹). R^2 values are calculated using only samples that had measureable ACE.

Journal of Environmental Quality

aquifer scale. However, the widespread occurrence of wastewater-affected groundwater suggests that septic system nutrients should not immediately be discounted when evaluating nutrient sources (e.g., nitrate) in rural areas otherwise dominated by agricultural lands. Furthermore, septic systems might still be a significant source of groundwater nutrients on more local scales, such as for specific wells or seeps. Robertson et al. (2016a) used total inorganic nitrogen/ACE and Cl⁻/ACE ratios to calculate that, in three Canadian watersheds dominated by agriculture, <15% of the groundwater nitrate could have potentially been derived from septic system effluents. The results presented here support the findings of previous studies that have used different methods to conclude that agricultural fertilizers are the main source of nitrate in the LASA (Hill 1982, 1986; Senger 2016).

Conclusions

Given that 11% of the Canadian population obtains their domestic water from private wells, and that these same households usually also have septic systems, the occurrence of artificial sweeteners in groundwater is likely widespread in rural areas across the country. In rural groundwater, septic systems are expected to be the major source of artificial sweeteners to groundwater, and therefore artificial sweeteners are a potential way of apportioning nutrient contributions between agricultural and septic sources, especially when used in combination with other novel tracers such as stable isotope ratios.

Although the Nottawasaga River has several municipal wastewater discharges, we show that groundwater discharging directly to the Nottawasaga River is also a source of artificial sweeteners to the river. Therefore, surface water bodies (rivers, streams, lakes) that do not have municipal wastewater discharges can still contain artificial sweeteners derived from groundwater affected by septic system effluents.

Given that ACE is so recalcitrant and mobile in groundwater, it is possible that even after other wastewater compounds such as pathogens and pharmaceutical compounds are completely removed, the presence of septic-derived water could still be detected using ACE. If ACE is not detected in a water sample, then other human wastewater-derived contaminants are also unlikely to be present at levels of concern.

Acknowledgments

We gratefully acknowledge the support of the land owners in the southern Nottawasaga River Watershed that let us collect samples of their well water. We thank Sue Brown, Eric Westberg, John Voralek, and Braden Kralt for assistance with sample collection and/or processing. Ryan Post, Nottawasaga Valley Conservation Authority, provided liaison support for the project. Stephanie McPhie, Nottawasaga Valley Conservation Authority, provided GIS support and prepared Fig. 1. Dr. Allan Hill, York University, provided information from his previous work in the study area that helped with the design of this study. We thank the anonymous reviewers for their suggestions that helped improve previous versions of this manuscript. Funding for the study was provided by Environment and Climate Change Canada, the Ontario Ministry of Agriculture Food and Rural Affairs, and the Natural Sciences and Engineering Research Council.

References

Barnes, K.K., D.W. Kolpin, E.T. Furlong, S.D. Zaugg, M.T. Meyer, and L.B. Barber. 2008. A national reconnaissance of pharmaceuticals and other organic wastewater contaminants in the United States: I) Groundwater. Sci. Total Environ. 402:192–200. doi:10.1016/j.scitotenv.2008.04.028

- Berger, B.M., and N.L. Wolfe. 1996. Multiresidue determination of sulfonylurea herbicides by capillary electrophoresis for hydrolysis studies in water and sediments. Fresenius J. Anal. Chem. 356:508–511.
- Brorström-Lundén, E., A. Svenson, T. Viktor, A. Woldegiorgis, M. Remberger, L. Kaj et al. 2008. Measurements of sucralose in the Swedish Screening Program 2007, PART I: Sucralose in surface waters and STP samples. Rep. B1769. Swedish Environ. Res. Inst., Stockholm, Sweden.
- Buerge, I.J., H.-R. Buser, M. Kahle, M.D. Müller, and T. Poiger. 2009. Ubiquitous occurrence of the artificial sweetener acesulfame in the aquatic environment: An ideal chemical marker of domestic wastewater in groundwater. Environ. Sci. Technol. 43:4381–4385. doi:10.1021/es900126x
- Buerge, I.J., M. Keller, H.-R. Buser, M.D. Müller, and T. Poiger. 2011. Saccharin and other artificial sweeteners in soils: Estimated inputs from agriculture and households, degradation, and leaching to groundwater. Environ. Sci. Technol. 45:615–621. doi:10.1021/es1031272
- Carrara, C., C.J. Ptacek, W.D. Robertson, D.W. Blowes, M.C. Moncur, E.D. Sverko, and S. Backus. 2008. Fate of pharmaceutical and trace organic compounds in three septic system plumes, Ontario, Canada. Environ. Sci. Technol. 42:2805–2811. doi:10.1021/es070344q
- Castronovo, S., A. Wick, M. Scheurer, K. Nödler, M. Schulz, and T.A. Ternes. 2017. Biodegradation of the artificial sweetener acesulfame in biological wastewater treatment and sandfilters. Water Res. 110:342–353. doi:10.1016/j.watres.2016.11.041
- Clarke, B.O., T. Anumol, M. Barlaz, and S.A. Snyder. 2015. Investigating landfill leachate as a source of trace organic pollutants. Chemosphere 127:269– 275. doi:10.1016/j.chemosphere.2015.02.030
- Drewes, J.E., T. Heberer, and K. Reddersen. 2002. Fate of pharmaceuticals during indirect potable reuse. Water Sci. Technol. 46:73–80.
- Ens, W., F. Senner, B. Gygax, and G. Schlotterbeck. 2014. Development, validation, and application of a novel LC-MS/MS trace analysis method for the simultaneous quantification of seven iodinated X-ray contrast media and three artificial sweeteners in surface, ground, and drinking water. Anal. Bioanal. Chem. 406:2789–2798. doi:10.1007/s00216-014-7712-0
- Environment and Climate Change Canada. 2017. Canadian climate normals 1981-2010 station data, Egbert, ON. Environ. Clim. Change Canada. http://climate.weather.gc.ca/climate_normals/index_e.html (accessed 8 June 2017).
- Foolad, M., S.L. Ong, and J. Hu. 2015. Transport of sewage molecular markers through saturated soil column and effect of easily biodegradable primary substrate on their removal. Chemosphere 138:553–559. doi:10.1016/j. chemosphere.2015.07.008
- Fram, M.S., and K. Belitz. 2011. Occurrence and concentrations of pharmaceutical compounds in groundwater used for public drinking-water supply in California. Sci. Total Environ. 409:3409–3417. doi:10.1016/j.scitotenv.2011.05.053
- Francy, D.S., D.R. Helsel, and R.A. Nally. 2000. Occurrence and distribution of microbiological indicators in groundwater and stream water. Water Environ. Res. 72:152–161. doi:10.2175/106143000X137220
- Gan, Z., H. Sun, B. Feng, R. Wang, and Y. Zhang. 2013. Occurrence of seven artificial sweeteners in the aquatic environment and precipitation of Tianjin, China. Water Res. 47:4928–4937. doi:10.1016/j.watres.2013.05.038
- Gan, Z., H. Sun, R. Wang, H. Hu, and P. Zhang. 2014. Transformation of acesulfame in water under natural sunlight: Joint effect of photolysis and biodegradation. Water Res. 64:113–122. doi:10.1016/j.watres.2014.07.002
- Goss, M.J., D.A.J. Barry, and D.L. Rudolph. 1998. Contamination in Ontario farmstead domestic wells and its association with agriculture: 1. Results from drinking water wells. J. Contam. Hydrol. 32:267–293. doi:10.1016/ S0169-7722(98)00054-0
- Gougeon, R., M. Spidel, K. Lee, and C.J. Field. 2004. Canadian diabetes association national nutrition committee technical review: Non-nutritive intense sweeteners in diabetes management. Can. J. Diabetes 28:385–399.
- Hill, A.R. 1982. Nitrate distribution in the ground water of the Alliston Region of Ontario, Canada. Ground Water 20:696–702. doi:10.1111/j.1745-6584.1982.tb01389.x
- Hill, A.R. 1986. Nitrate and chloride distribution and balance under continuous potato cropping. Agric. Ecosyst. Environ. 15:267–280. doi:10.1016/0167-8809(86)90125-8
- Khan, I.U., V. Gannon, R. Kent, W. Koning, D.R. Lapen, J. Miller et al. 2007. Development of a rapid quantitative PCR assay for direct detection and quantification of culturable and non-culturable *Escherichia coli* from agriculture watersheds. J. Microbiol. Methods 69:480–488. doi:10.1016/j.mimet.2007.02.016
- Lange, F.T., M. Scheurer, and H.J. Brauch. 2012. Artificial sweeteners—A recently recognized class of emerging environmental contaminants: A review. Anal. Bioanal. Chem. 403:2503–2518. doi:10.1007/s00216-012-5892-z
- Lee, D.G., P.R. Roehrdanz, M. Feraud, J. Ervin, T. Anumol, A. Jia et al. 2015. Wastewater compounds in urban shallow groundwater wells correspond to exfiltration probabilities of nearby sewers. Water Res. 85:467–475. doi:10.1016/j.watres.2015.08.048

- Li, A.J., O.J. Schmitz, S. Stephan, C. Lenzen, P.Y.K. Yue, K. Li et al. 2016. Photocatalytic transformation of acesulfame: Transformation products identification and embryotoxicity study. Water Res. 89:68–75. doi:10.1016/j. watres.2015.11.035
- Maupin, M.A., J.F. Kenny, S.S. Hutson, J.K. Lovelace, N.L. Barber, and K.S. Linsey. 2014. Estimated use of water in the United States in 2010. Circ. 1405. USGS, Reston, VA. doi:10.3133/cir1405.
- Nödler, K., O. Hillebrand, K. Idzik, M. Strathmann, F. Schiperski, J. Zirlewagen, and T. Licha. 2013. Occurrence and fate of the angiotensin II receptor antagonist transformation product valsartan acid in the water cycle: A comparative study with selected β-blockers and the persistent anthropogenic wastewater indicators carbamazepine and acesulfame. Water Res. 47:6650–6659. doi:10.1016/j.watres.2013.08.034
- Nottawasaga Valley Conservation Authority. 2013. 2013 Nottawasaga Valley Watershed health check. Nottawasaga Valley Conserv. Auth. http://www. nvca.on.ca/Shared Documents/2013 NVCA WHC.pdf (accessed 11 Jan. 2016).
- Ontario Ministry of Agriculture, Food and Rural Affairs. 2016a. Western Ontario region at a glance. Ontario Minist. Agric. http://www.omaf.gov.on.ca/ english/stats/county/western ontario.xls (accessed on 17 Feb. 2016).
- Ontario Ministry of Agriculture, Food and Rural Affairs. 2016b. Number of pigs, by county, July 2014. Ontario Minist. Agric. http://www.omaf.gov. on.ca/english/stats/livestock/ctypigs14.htm (accessed on 17 Feb. 2016).
- Oppenheimer, J., A. Eaton, M. Badruzzaman, A.W. Haghani, and J.G. Jacangelo. 2011. Occurrence and suitability of sucralose as an indicator compound of wastewater loading to surface waters in urbanized regions. Water Res. 45:4019–4027. doi:10.1016/j.watres.2011.05.014
- Pasquini, L., J.-F. Munoz, N. Rimlinger, X. Dauchy, X. France, M.-N. Pons, and T. Görner. 2013. Assessment of the fate of some household micropollutants in urban wastewater treatment plant. Chem. Pap. 67:601–612. doi:10.2478/s11696-013-0339-3
- Prasse, C., M. Wagner, R. Schulz, and T.A. Ternes. 2011. Biotransformation of the antiviral drugs acyclovir and penciclovir in activated sludge treatment. Environ. Sci. Technol. 45:2761–2769. doi:10.1021/es103732y
- Renwick, A.G. 1986. The metabolism of intense sweeteners. Xenobiotica 16:1057–1071. doi:10.3109/00498258609038983
- Robertson, W.D., D.R. Van Stempvoort, J.W. Roy, S.J. Brown, J. Spoelstra, S.L. Schiff et al. 2016a. Use of an artificial sweetener to identify sources of groundwater nitrate contamination. Ground Water. doi:10.1111/gwat.12399
- Robertson, W.D., D.R. Van Stempvoort, D.K. Solomon, J. Homewood, S.J. Brown, J. Spoelstra, and S.L. Schiff. 2013. Persistence of artificial sweeteners in a 15-year-old septic system plume. J. Hydrol. 477:43–54. doi:10.1016/j.jhydrol.2012.10.048
- Robertson, W.D., D.R. Van Stempvoort, J. Spoelstra, S.J. Brown, and S.L. Schiff. 2016b. Degradation of sucralose in groundwater and implications for age dating contaminated groundwater. Water Res. 88:653–660. doi:10.1016/j. watres.2015.10.051
- Roslev, P., and A.S. Bukh. 2011. State of the art molecular markers for fecal pollution source tracking in water. Appl. Microbiol. Biotechnol. 89:1341–1355. doi:10.1007/s00253-010-3080-7
- Roy, J.W., D.R. Van Stempvoort, and G. Bickerton. 2014. Artificial sweeteners as potential tracers of municipal landfill leachate. Environ. Pollut. 184:89– 93. doi:10.1016/j.envpol.2013.08.021
- Rusiñol, M., X. Fernandez-Cassi, A. Hundesa, C. Vieira, A. Kern, I. Eriksson et al. 2014. Application of human and animal viral microbial source tracking tools in fresh and marine waters from five different geographical areas. Water Res. 59:119–129. doi:10.1016/j.watres.2014.04.013
- Sang, Z., Y. Jiang, Y.K. Tsoi, and K.S.-Y. Leung. 2014. Evaluating the environmental impact of artificial sweeteners: A study of their distributions, photodegradation and toxicities. Water Res. 52:260–274. doi:10.1016/j. watres.2013.11.002
- Schaider, L.A., J.M. Ackerman, and R.A. Rudel. 2016. Septic systems as sources of organic wastewater compounds in domestic drinking water wells in a shallow sand and gravel aquifer. Sci. Total Environ. 547:470–481. doi:10.1016/j.scitotenv.2015.12.081
- Schaider, L.A., R.A. Rudel, J.M. Ackerman, S.C. Dunagan, and J. Green Brody. 2014. Pharmaceuticals, perflorosurfactants, and other organic wastewater compounds in public drinking water wells in a shallow sand and gravel aquifer. Sci. Total Environ. 468–469:384–393. doi:10.1016/j. scitotenv.2013.08.067
- Scheurer, M., H.-J. Brauch, and F.T. Lange. 2009. Analysis and occurrence of seven artificial sweeteners in German waste water and surface water and in soil aquifer treatment (SAT). Anal. Bioanal. Chem. 394:1585–1594. doi:10.1007/s00216-009-2881-y

- Scheurer, M., M. Godejohann, A. Wick, O. Happel, T.A. Ternes, H.J. Brauch et al. 2012. Structural elucidation of main ozonation products of the artificial sweeteners cyclamate and acesulfame. Environ. Sci. Pollut. Res. 19:1107– 1118. doi:10.1007/s11356-011-0618-x
- Scheurer, M., B. Schmutz, O. Happel, H.-J. Brauch, R. Wülser, and F.R. Storck. 2014. Transformation of the artificial sweetener acesulfame by UV light. Sci. Total Environ. 481:425–432. doi:10.1016/j.scitotenv.2014.02.047
- Scheurer, M., F.R. Storck, H.-J. Brauch, and F.T. Lange. 2010. Performance of conventional multi-barrier drinking water treatment plants for the removal of four artificial sweeteners. Water Res. 44:3573–3584. doi:10.1016/j. watres.2010.04.005
- Seiler, R.L., S.D. Zaugg, J.M. Thomas, and D.L. Howcroft. 1999. Caffeine and pharmaceuticals as indicators of waste water contamination in wells. Ground Water 37:405–410. doi:10.1111/j.1745-6584.1999.tb01118.x
- Senger, N.D. 2016. Multi-decade comparison of groundwater nitrate in the southern Nottawasaga River Watershed. Master's thesis. Univ. of Waterloo, Waterloo, ON. https://uwspace.uwaterloo.ca/handle/10012/10693 (accessed 6 Sept. 2016).
- Sibul, U., and A.V. Choo-Ying. 1971. Water Resources of the Upper Nottawasaga River Drainage Basin. Water Resour. Rep. 3. Ontario Water Resour. Comm., Toronto, ON.
- Spoelstra, J., S.L. Schiff, and S.J. Brown. 2013. Artificial sweeteners in a large Canadian river reflect human consumption in the watershed. PLoS One 8:e82706. doi:10.1371/journal.pone.0082706 [erratum: 9:e106394].
- Statistics Canada. 2011. Households and the environment. Catalogue no. 11-526-X. Statistics Canada. http://www.statcan.gc.ca/pub/11-526-x/11-526-x2013001-eng.htm (accessed 7 Jan. 2016).
- Subedi, B., S. Lee, H.-B. Moon, and K. Kannan. 2014. Emission of artificial sweeteners, select pharmaceuticals, and personal care products through sewage sludge from wastewater treatment plants in Korea. Environ. Int. 68:33–40. doi:10.1016/j.envint.2014.03.006
- Tran, N.H., J. Gan, V.T. Nguyen, H. Chen, L. You, A. Duarah et al. 2015. Sorption and biodegradation of artificial sweeteners in activated sludge processes. Bioresour. Technol. 197:329–338. doi:10.1016/j.biortech.2015.08.083
- Tran, N.H., V.T. Nguyen, T. Urase, and H.H. Ngo. 2014. Role of nitrification in the biodegradation of selected artificial sweetening agents in biological wastewater treatment process. Bioresour. Technol. 161:40–46. doi:10.1016/j.biortech.2014.02.116
- Van Stempvoort, D.R., W.D. Robertson, and S. Brown. 2011a. Artificial sweeteners in a large septic plume. Groundwater Monit. Rem. 31:95–102. doi:10.1111/j.1745-6592.2011.01353.x
- Van Stempvoort, D.R., J.W. Roy, S.J. Brown, and G. Bickerton. 2011b. Artificial sweeteners as potential tracers in groundwater in urban environments. J. Hydrol. 401:126–133. doi:10.1016/j.jhydrol.2011.02.013
- Van Stempvoort, D.R., J.W. Roy, J. Grabuski, S.J. Brown, G. Bickerton, and E. Sverko. 2013. An artificial sweetener and pharmaceutical compounds as co-tracers of urban wastewater in groundwater. Sci. Total Environ. 461– 462:348–359. doi:10.1016/j.scitotenv.2013.05.001
- Van Stempvoort, D.R., Spoelstra, J., Senger, N.D., Brown, S.J., Post, R., Struger, J. 2016. Glyphosate residues in rural groundwater, Nottawasaga River Watershed, Ontario, Canada. Pest Manage. Sci. 72:1862–1872. doi:10.1002/ ps.4218
- Withers, P.J.A., P. Jordan, L. May, H.P. Jarvie, and N.E. Deal. 2014. Do septic tank systems pose a hidden threat to water quality? Front. Ecol. Environ 12:123–130. doi:10.1890/130131
- Wolf, L., C. Zwiener, and M. Zemann. 2012. Tracking artificial sweeteners and pharmaceuticals introduced into urban groundwater by leaking sewer networks. Sci. Total Environ. 430:8–19. doi:10.1016/j. scitotenv.2012.04.059
- Wu, M., Y. Qian, J.M. Boyd, S.E. Hrudey, X.C. Le, and X.F. Li. 2014. Direct large volume injection ultra-high performance liquid chromatography-tandem mass spectrometry determination of artificial sweeteners sucralose and acesulfame in well water. J. Chromatogr. A 1359:156–161. doi:10.1016/j. chroma.2014.07.035
- Yang, Y.Y., W.R. Liu, Y.S. Liu, J.L. Zhao, Q.Q. Zhang, M. Zhang et al. 2017. Suitability of pharmaceuticals and personal care products (PPCPs) and artificial sweeteners (ASs) as wastewater indicators in the Pearl River Delta, South China. Sci. Total Environ. 590–591:611–619. doi:10.1016/j. scitotenv.2017.03.001
- Zirlewagen, J., T. Licha, F. Schiperski, K. Nödler, and T. Scheytt. 2016. Use of two artificial sweeteners, cyclamate and acesulfame, to identify and quantify wastewater contributions in a karst spring. Sci. Total Environ. 547:356– 365. doi:10.1016/j.scitotenv.2015.12.112