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Artificial Sweetener Sucralose in U.S. Drinking Water Systems

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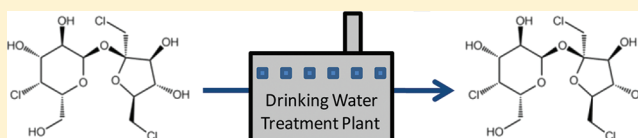
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S Supporting Information

ABSTRACT:



The artificial sweetener sucralose has recently been shown to be a widespread contaminant of wastewater, surface water, and groundwater. In order to understand its occurrence in drinking water systems, water samples from 19 United States (U.S.) drinking water treatment plants (DWTPs) serving more than 28 million people were analyzed for sucralose using liquid chromatography tandem mass spectrometry (LC-MS/MS). Sucralose was found to be present in source water of 15 out of 19 DWTPs (47–2900 ng/L), finished water of 13 out of 17 DWTPs (49–2400 ng/L) and distribution system water of 8 out of the 12 DWTPs (48–2400 ng/L) tested. Sucralose was only found to be present in source waters with known wastewater influence and/or recreational usage, and displayed low removal (12% average) in the DWTPs where finished water was sampled. Further, in the subset of DWTPs with distribution system water sampled, the compound was found to persist regardless of the presence of residual chlorine or chloramines. In order to understand intra-DWTP consistency, sucralose was monitored at one drinking water treatment plant over an 11 month period from March 2010 through January 2011, and averaged 440 ng/L in the source water and 350 ng/L in the finished water. The results of this study confirm that sucralose will function well as an indicator compound for anthropogenic influence on source, finished drinking and distribution system (i.e., tap) water, as well as an indicator compound for the presence of other recalcitrant compounds in finished drinking water in the U.S.

INTRODUCTION

Artificial sweeteners are a class of organic contaminants recently discovered in wastewater, groundwater, surface water, bank filtrate and drinking water.^{1–7} These compounds are widely added to foods, personal care products and pharmaceutical formulations, and enter the environment in part due to incomplete removal during wastewater treatment. While artificial sweeteners undergo comprehensive toxicological testing prior to their use in consumer products, their unintended presence in water is still cause for concern. This is because the long-term health effects resulting from chronic exposure to low levels of these compounds and other trace organic contaminants in water, such as pharmaceuticals and personal care products,^{8,9} are largely unknown.^{10,11} However, the true importance of the artificial sweeteners will likely be as indicator compounds for wastewater influence on other water types.^{1,3,6} In any case, the quantification of these compounds in various water types, including finished drinking water, is the first step in understanding the significance of this issue.

A great deal of analytical work for artificial sweeteners has concerned measuring their concentration in food products.^{12–17} However, there are a growing number of studies of artificial

sweetener occurrence in wastewater and surface water. A large volume of this work has been completed in Europe, where acesulfame and sucralose were found to be the most persistent through wastewater treatment and in the environment.^{1,3,4} Acesulfame was found to occur at higher levels than sucralose in the waste-, surface, ground, and tap water of Switzerland in Buerge et al.¹ and wastewaters and surface waters of Germany in Scheurer et al.³ Both studies proposed acesulfame as an indicator compound for domestic wastewater influence on other water types. The occurrence of sucralose in surface waters has been measured in 27 European countries in Loos et al.,⁴ as well as smaller studies in Switzerland¹ and Germany³ that included groundwater. This compound has also been proposed as an indicator compound of domestic wastewater influence on surface water,³ although acesulfame was found to be the predominant artificial sweetener in these studies. Both compounds were the subject of a recent study of bench-scale and full-scale drinking

Received: July 12, 2011

Accepted: August 31, 2011

Revised: August 29, 2011

Published: August 31, 2011

Table 1. Summary of Treatment Processes Employed at the DWTPs Included in This Study (Adapted From Ref ⁹) and the Concentration of Sucralose Determined in the Source, Finished and Distribution System Water for Each Site^a

DWTP No.	treatment scheme	influence	source (ng/L)	finished (ng/L)	% removal	distribution (ng/L)
1	C/F → Sed. → Cl ₂ → Filt. → NH ₂ Cl	ww	2400	2400	0	2400
2	ClO ₂ → C/F → Sed. → Filt. → UV → Cl ₂	ww	2900	1500	48	n/a
3	ClO ₂ → C/F → Sed. → Filt. → UV → Cl ₂	ww	790	1100 ^b (680)	14	1100 ^b (680)
4	Cl ₂ → C/F → Sed. → Cl ₂ → Filt. → Cl ₂	ww	220	210	5	200
5	Cl ₂ → O ₃ → C/F → Filt. → Cl ₂	ww	240	n/a	n/a	n/a
6	Cl ₂ → O ₃ → C/F → Filt. → Cl ₂	ww	240	190	21	180
7	Cl ₂ → C/F → Sed. → Cl ₂ → Filt. → NH ₂ Cl	ww	2200	2000	9	n/a
8	Cl ₂ → C/F → Sed. → Cl ₂ → Filt. → NH ₂ Cl	ww	1500	1500	0	n/a
9	Cl ₂ → C/F → Sed. → Cl ₂ → Filt. → NH ₂ Cl	ww	400	370	8	n/a
10	Cl ₂ → C/F → Sed. → Filt. → NH ₂ Cl	n	<MRL	<MRL	n/a	<MRL
11	Cl ₂ → C/F → Sed. → Filt. → NH ₂ Cl	n	<MRL	<MRL	n/a	<MRL
12	Cl ₂ → NH ₂ Cl	n	<MRL	<MRL	n/a	<MRL
13	Cl ₂ → O ₃ → C/F → Sed. → NH ₂ Cl	ww	150	130	13	120
14	Filt. → O ₃ → NH ₂ Cl	n	<MRL	<MRL	n/a	<MRL
15	Cl ₂ → C/F → Sed. → Cl ₂ → Filt. → NH ₂ Cl	ww	100	n/a	n/a	93
16	C/F → Sed. → O ₃ → dm-Filt. → NH ₂ Cl	r	81	55	32	53
17	Cl ₂ → C/F → Sed. → Cl ₂ → Filt. → NH ₂ Cl	r	47	49	(+4%)	48
18	O ₃ → C/F → Sed. → GAC/sand bio-Filt. → NH ₂ Cl	r	72	108	(+50%)	n/a
19	O ₃ → C/F → Sed. → GAC/sand bio-Filt. → NH ₂ Cl	r	62	98	(+58%)	n/a

^a C/F: coagulation/flocculation; Sed.: sedimentation; Cl₂: free-chlorine (hypochlorite); Filt.: filtration; NH₂Cl: chloramine; ClO₂: chlorine dioxide; pre-ox.: O₃; ozonation; dm-Filt.: dual-media filtration; GAC/sand bio-Filt.: granular activated carbon/sand biofiltration; ww: wastewater; n: none; r: recreational; <MRL: below method reporting limit; n/a: not available for analysis. ^b The finished water for DWTP-3 is a blend, and the value in parentheses has a blending factor applied.

water treatment for their removal, where acesulfame was found to somewhat persistent, while sucralose was effectively removed by granular activated carbon in Scheurer et al.⁷

There is a growing body of work related to the artificial sweetener sucralose in the United States and Canada. Sucralose was measured in the marine and coastal waters of the U.S., as well as wastewater treatment plant effluent and river water subject to its discharge, where concentrations reached 1.9 µg/L in Mead et al.² Later, sucralose was detected in five out of five wastewater samples from three locations, at a median concentration of 1.5 µg/L in Ferrer and Thurman.⁵ It was also detected in eight out of 22 surface water samples, in eight out of eight alluvial groundwater samples from two locations, and suggested as a conservative tracer of sewage effluent in surface and groundwater. Most recently, sucralose was demonstrated to be a viable indicator compound for wastewater loading in surface waters, where it was consistently found in wastewater, wastewater-influenced surface water and septic samples in Oppenheimer et al.⁶ Sucralose occurrence has also recently been measured in urban groundwater in Canada, although acesulfame was found more consistently in Van Stempvoort et al.¹⁸ Sucralose has been found to be persistent through wastewater treatment processes in municipal plants in Torres et al.,¹⁹ and some bench scale studies in Soh et al.²⁰

Sucralose, a chlorinated form of sucrose, is an artificial sweetener that is approximately 600 times sweeter than sucrose and is widely used as a food additive.²¹ It is not extensively adsorbed and metabolized in humans, resulting in the majority being excreted without transformation.²² Sucralose is very soluble in water (282 g/L at 20 °C, Log *P* = −0.51),²³ so it is not expected to be extensively adsorbed to organic solids in the environment. Further, sucralose has been shown to be resistant

to treatment processes used in the production of drinking water at the bench scale, such as oxidation by free chlorine and ozone.²⁰ Because of these properties, it is likely that sucralose may be widespread in drinking waters in the U.S., persisting through traditional drinking water treatment processes.

The objective of this work was to quantify the amount of sucralose present in select U.S. drinking water systems. To this end, an archived set of extracts from the study reported in Benotti et al.,⁹ of the source, finished, and distribution system water from 19 U.S. DWTPs from 2006 to 2007, was analyzed for this compound. The relative ineffectiveness of commonly employed drinking water treatment strategies to remove sucralose is presented. The utility of this compound as an indicator for anthropogenic influence on drinking water systems, and as an indicator for other recalcitrant compounds in finished drinking water, is also discussed. To date, this manuscript represents the first study to quantify the artificial sweetener sucralose in U.S. drinking water systems.

EXPERIMENTAL SECTION

Sample Collection. The sampling sites consisted of the source, finished and distribution systems of 19 U.S. DWTPs. This is the same sample set analyzed by Benotti et al.⁹ Only 10 of the 19 DWTPs had sites in the distribution system sampled, and one finished water sample extract from one DWTP was lost during storage and handling. The treatment process employed at each plant is summarized in Table 1. The hydraulic retention time was taken into account when collecting samples so that the data should represent, as closely as possible, the same plug of water moving through the treatment process.

Samples were collected in 1 L silanized, baked amber glass bottles containing the preservative sodium azide and ascorbic acid to quench any residual oxidant. All samples were cooled on ice, shipped overnight and stored at 4 °C in the laboratory for a maximum of 14 days until extraction. Travel blanks were included with each sampling event, and consisted of laboratory-grade water that was shipped with the bottles, and transferred to a sampling bottle during the sampling procedure. All samples were extracted using the SPE method described in Vanderford et al.,²⁴ and resulted in a 1000-fold concentration. Following extraction, the sample extracts in methanol were capped in airtight sample vials and stored at -80 °C until the time of analysis, which totaled 36–48 months depending on sampling and extraction dates. Prior to analysis, extracts were allowed to warm to room temperature and a positive displacement pipet was used to transfer 40 μ L to an autosampler vial containing 60 μ L of a solution of the isotope-labeled internal standard, sucralose-*d*₆ (Toronto Research Chemicals, Toronto, ON). After thorough mixing using a glass pipet, the sample was diluted 2.5 times to a final composition of 46% MeOH, 54% DI water, and an internal standard concentration of 200 ng/mL. The original extract was capped and returned to the -80 °C freezer, and the prepared extract was stored at 4 °C until analysis.

The source waters of the DWTPs included in this study display a range of anthropogenic influence. Only one of the DWTPs utilizes a groundwater source (DWTP-12), while the remaining employ surface water. The source water from DWTPs 1 to 9, 13, and 15 are impacted by wastewater inputs. The source waters for DWTPs 16 to 19 are reservoirs with recreational usage, but no known wastewater inputs. The remaining DWTPs have no known wastewater inputs or recreational usage.

Analysis Method. All extracts were analyzed by isotope dilution liquid chromatography–tandem mass spectrometry (LC-MS/MS). The chromatography was performed on an Agilent (Santa Clara, CA) 1200 system equipped with a Restek (Bellefonte, PA) Allure Organic Acids column, 150 \times 3.2 mm with 5 μ m particles. Mass spectrometric analysis was performed using an AB SCIEX (Foster City, CA) API-4000 QTrap operating with negative electrospray ionization in MRM mode. Sucralose was quantified using the (M-H)⁻ precursor ion at *m/z* 395.0 and the Cl⁻ product ion at *m/z* 35.0 and confirmed using the (M-H)⁻ precursor ion at *m/z* 397.0 and the Cl⁻ product ion at *m/z* 35.0. The internal standard was measured using the (M-H)⁻ precursor ion at *m/z* 403.0 and the Cl⁻ product ion at *m/z* 35.0 in order to avoid significant contribution from the naturally occurring isotopes of sucralose. A product ion spectrum of the precursor ion at *m/z* 395.0 collected at a collision energy of 22 eV is shown in Figure 1. Further details on the analytical method can be found in the Supporting Information. Representative LC-MS/MS chromatograms for source water, finished water and travel blank extracts from a sampling event for DWTP-8 can also be found in the Supporting Information (Figure SI-1).

RESULTS AND DISCUSSION

Analytical Parameters and Data Quality. The percent recovery was determined by analyzing 10 samples of laboratory reagent water spiked with 250 ng/L of sucralose and with 200 ng/L of isotopically labeled internal standard, taken through the entire sample extraction procedure. The absolute recovery of sucralose was determined by external calibration (i.e., - no internal standard correction) to be 85 \pm 7%. This represents the

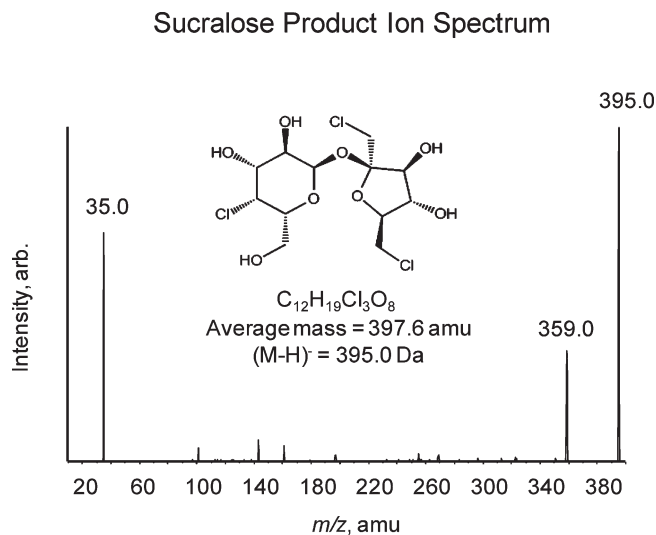


Figure 1. Sucralose product ion spectrum in negative ion mode with a collision energy of -22 V. The molecular structure, empirical formula, and average mass are included. The precursor ion is the (M-H)⁻ species at *m/z* = 395.0 Da.

recovery of the sucralose in the samples through the extraction procedure. Relative recovery was determined by ratioing the sucralose to the internal standard in order to account for any analyte ionization suppression and extraction efficiency, which was calculated to be 99%. Since the internal standard was added postextraction in the procedure used for the archived extracts, this only corrects for ion suppression, but not for the extraction recovery. The results reported in this manuscript are reported as such, with no correction for extraction efficiency. It should be noted that the artificial sweeteners saccharin and acesulfame were also considered for analysis; however, the extraction procedure used in the original study⁹ resulted in very low recoveries. Therefore, they could not be quantified in these extracts.

The minimum detection limit (MDL) was determined from the analysis of eight samples of laboratory reagent water spiked with 10 ng/L of sucralose and 200 ng/L of the internal standard. The samples were subjected to the entire sample preparation procedure and quantified by the LC-MS/MS method using internal calibration. The MDL was calculated from the resulting data by multiplying the standard deviation of these eight replicate measurements by the appropriate Student's *T*-value for *n* - 1 degrees of freedom, and determined to be 1.6 ng/L. The low calibrator and reporting limit was set to a factor 6 times higher, or 10 ng/L. A similar procedure has been used previously to determine MDL values for pharmaceutical compounds in water matrices in Vanderford et al.²⁴

Because the analyses were performed on a set of sample extracts archived for 36–48 months at -80 °C, there are concerns with sample integrity during long-term storage. Sucralose has been shown to be a relatively inert and stable molecule, and is known to be very soluble in the storage solvent, methanol.²⁵ There were several duplicate samples collected throughout the study for source, finished drinking and distribution water. The majority differed by less than 10% of the average of the two values, although one distribution duplicate sample had a 15% difference. This error is well within that expected in typical water analysis methods, and indicates acceptable data quality. It also shows that the long-term storage conditions did not cause

significant differences between samples. Additionally, sucralose was not detected in any of the travel blanks, demonstrating the integrity of sample collection and handling procedures.

Sucralose Occurrence in Source and Finished Water. Sucralose was detected in the majority of source water samples, and displayed a strong dependence on the relative amount of anthropogenic influence. Those source waters that were impacted by wastewater all tested positive and displayed the highest concentrations, ranging from 100 ng/L (DWTP-15) to 2900 ng/L (DWTP-2), as shown in Table 1. Source waters that had no known wastewater inputs but were subject to recreational usage, such as boating and swimming, also tested positive but displayed lower concentrations than those impacted by wastewater. This sample group also displayed very similar levels to one another, ranging from 47 (DWTP-17) to 81 ng/L (DWTP-16), with sucralose likely being present from improper disposal of food-stuffs and human waste.²² Those source waters that had no known wastewater inputs or recreational usage did not contain quantifiable levels of sucralose (DWTP-10 to 12, 14).

The concentrations reported in this work are comparable with previously published levels of artificial sweeteners^{2–6} in surface and groundwater. Sucralose concentrations range higher than those reported in European surface waters,^{3,4} but are comparable to those measured in American surface and ground waters,^{2,5,6} likely due to the time period for its approved usage (beginning in 1998 in the U.S. versus 2004 in the E.U.), as well as differences in artificial sweetener usage patterns. For results obtained in U.S. waters, the concentrations reported here range up to 2900 ng/L, compared with 1900 ng/L by Mead et al., 2400 ng/L by Ferrer and Thurman, and 10,000 ng/L by Oppenheimer et al. Interestingly, these results are from surface water samples, with the exception of Ferrer et al., which is from a groundwater sample, demonstrating that this compound can be of concern even for those DWTPs with groundwater sources.

All of the DWTPs that used source water that tested positive for sucralose also had quantifiable amounts in the finished water, with the highest level at 2400 ng/L (DWTP-1). In fact, most DWTPs showed very little removal, including those that employed ozone as an oxidant. Counting any increases in concentration between finished and source water as 0% removal, the average removal across the plants with representative samples was 12%. DWTP-2 displayed the highest removal (48%), and employed ClO₂ preoxidation and UV treatment prior to chlorination. Although DWTP-3 has significantly less removal (14%), this still suggests that advanced oxidation processes (AOP) should be explored further for the removal of sucralose from drinking water.

Persistence of Sucralose in Distribution Systems. The distribution system water of the DWTPs whose finished water tested positive for sucralose also tested positive (Figure 2). Further, within expected experimental error, every distribution system sample tested at the same levels as the finished water. These results indicate that residual free chlorine or chloramines are not effective at removing sucralose during transit from the DWTP to the distribution sampling point. As mentioned in the Experimental Section, these sampling sites were chosen to be relatively far removed from the DWTP, so it can be expected that sucralose persists completely through the distribution system.

Every distribution water (i.e., tap water) that was produced from source water with measurable amounts of sucralose, also had sucralose present (Table 1, Figure 2). Therefore, it is

Sucralose Levels in Source, Finished and Distribution Water

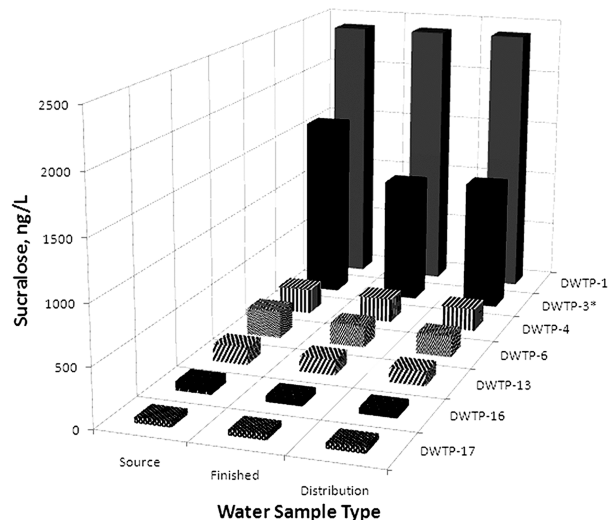


Figure 2. Comparison of sucralose concentration the source, finished and distribution water samples from seven utilities. These seven DWTPs represent those that had quantifiable amounts of sucralose in the source water, and had at least one finished and one distribution sample available for testing. *DWTP-3 was corrected for relative input to blend.

Sucralose in DWTP-5 Source and Finished Water and Comparison to Meprobamate Levels

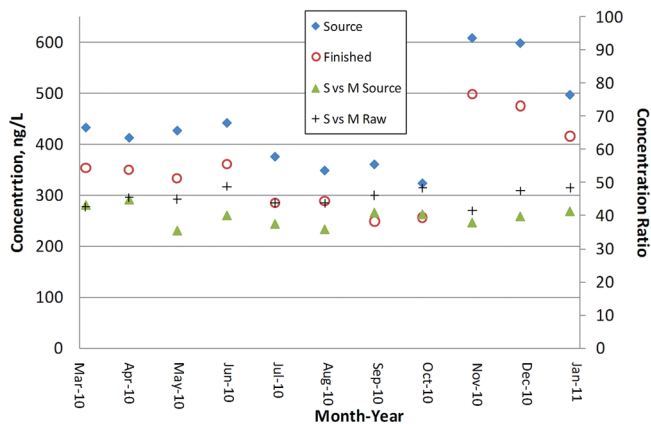


Figure 3. Sucralose concentrations in the source and finished water of DWTP-5 measured monthly by isotope dilution LC-MS/MS, and it is ratio to the meprobamate concentrations in the same sample sets. The average daily output of the DWTP during this time was 130 million gallons/day.

reasonable to expect that human exposure to sucralose via tap water is widespread in the U.S. Since the exact amount of tap water consumed daily on an individual basis varies, an average consumption amount of 2 L is estimated by the U.S. Environmental Protection Agency (US EPA).²⁶ Using this volume of tap water, the largest exposure to sucralose was from DWTP-1, where an individual would have been exposed to 4.8 μ g of sucralose per day. The toxicological relevance of chronic exposure to this amount warrants further studies, but is beyond the scope of this paper.

11-Month Monitoring Program at DWTP-5. In order to test whether the presence of sucralose is consistent over time, the

Table 2. Comparison of the Average Concentrations, Median Concentrations and Concentration Range of Sucralose with Compounds That Were Consistently Present for Each Water Type with Either Wastewater (ww) or Recreational Usage (r) Influence from Supporting Information in Benotti et al.⁹

water type	influence	analyte	average concentration, ng/L	median concentration (ng/L)	concentration range (ng/L)	MRL (ng/L)
source						
	ww	sucralose	1000	400	150–2900	10
	ww	carbamazepine	17	4.8	1.9–51	0.50
	ww	phenytoin	13	8.7	2.5–29	1.0
	ww	meprobamate	20	10	3.2–73	0.25
	ww	sulfamethoxazole	35	15	1.6–110	0.25
	ww	atrazine	180	32	1.4–870	0.25
	r	sucralose	66	67	47–81	10
	r	carbamazepine	1.5	1.7	0.60–1.8	0.50
	r	meprobamate	3.0	3.1	1.6–4.3	0.25
	r	sulfamethoxazole	1.7	1.6	0.62–3.2	0.25
	r	atrazine	310	230	0.50–780	0.25
finished						
	ww	sucralose	1000	1100	130–2400	10
	ww	phenytoin	8.7	6.2	1.1–19	1.0
	ww	meprobamate	17	13	3.3–42	0.25
	ww	atrazine	150	59	0.88–870	0.25
	r	sucralose	78	77	49–108	10
	r	meprobamate	1.4	1.5	1.0–1.8	0.25
	r	atrazine	120	110	0.38–240	0.25
distribution						
	ww	sucralose	680	190	93–2400	10
	ww	phenytoin	5.3	3.6	1.1–15	1.0
	ww	meprobamate	14	6.1	2.7–38	0.25
	ww	atrazine	186	113	0.94–915	0.25
	r	sucralose	51	51	48–53	10
	r	meprobamate	1.5	1.5	1.3–1.6	0.25
	r	atrazine	0.47	0.47	0.36–0.58	0.25

concentration in the raw and finished drinking water at DWTP-5 was monitored over the 11-month period from March, 2010 until January, 2011 (Figure 3). This DWTP has an ozone contact time of 24 min and a chlorine contact time of ~ 1.5 h. The amount of sucralose was quantified by isotope dilution LC-MS/MS using the method outlined in the Experimental Section, with the exception that the isotope was spiked into the sample prior to extraction, and the methanol extract was analyzed without dilution. This procedure corrects both for losses due to extraction and handling, as well as any ion suppression during analysis. The concentration of sucralose was relatively stable in both sample sets over that time, indicating that the flux of sucralose to that body of water was relatively consistent. There was an increase in concentration between October and November, which is due to seasonal fluctuations in the mixing dynamics of the treated wastewater stream and the reservoir, as determined previously through water quality parameters.^{27,28} The removal efficiency is consistently low through the treatment process (20% average removal), demonstrating the resistance of the compound to oxidation by both ozone and chlorine.

Sucralose as an Indicator Compound. Indicator compounds for drinking water have been defined as anthropogenic

compounds that can be used to represent certain classes of compounds based on their similarities in occurrence and behavior under various environmental and treatment conditions.²⁹ They have also been defined as chemical compounds that denote the presence of different types of water influence, usually wastewater.⁶ The data presented here confirm that sucralose is a reliable indicator compound for anthropogenic influence on source water across the United States. It is important to note that it is not only present in water that is directly impacted by wastewater, but also water where recreational use is permitted (with no known wastewater inputs). Given the poor removal of sucralose during the various treatment schemes, it is also a viable indicator compound in finished and distribution water for other resistant compounds, such as meprobamate and atrazine.^{9,30} In fact, when the concentration of sucralose is ratioed to that of meprobamate for data collected over a 11-month period at DWTP-5, relatively consistent values are obtained for both the source (average ratio = 41, %RSD = 8.4) and finished (average ratio = 46, %RSD = 5.7) water (Figure 3).

A comparison of the sucralose data with those from the much broader study by Benotti et al.⁹ reveals important trends. Table 2 shows the concentrations of the compounds that were

consistently present based on type (source, finished or distribution) and influence (wastewater or recreational usage). Sucralose, atrazine and meprobamate were the only analytes from this compound set that were present in source, finished and distribution water, regardless of the influence type. The source of sucralose and meprobamate is likely from direct human activities, whereas the majority of atrazine present is likely from agricultural application and runoff. Therefore, sucralose and meprobamate should represent better indicators of wastewater and/or direct human input to source water, and subsequently finished drinking and distribution (i.e., tap) waters. While the concentration of sucralose was much higher than the concentration of meprobamate for all of the waters tested, from an analytical standpoint, the concentrations are comparable in relation to the respective method reporting limits (MRLs). A comparison with the meprobamate data presented by Benotti et al.⁹ is included in the Supporting Information (Table SI-1). Given expected decreases in the input of meprobamate from human usage,^{31–33} sucralose will likely be the more important indicator compound in the future.

■ ASSOCIATED CONTENT

S Supporting Information. Additional information as noted in the text. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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■ ACKNOWLEDGMENT

We thank Janie Holady and Josephine Chu for their assistance throughout the project. The authors also thank the Water Research Foundation (Project No. 3085) for funding the initial sample collection and extraction. T.B. was funded by a National Science Foundation (NSF) CAREER Award (EAR 087683).

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