Investigation of spatial distributions and temporal trends of triclosan in Canadian surface waters

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Abstract

Triclosan is widely used in personal care products (skin creams, toothpastes, soaps, deodorants, body spray) and cleaning products (dishwashing detergent and all purpose cleaners) (Halden 2014). In 2001 it was selected for screening level risk assessment under the *Canadian Environmental Protection Act* (HC and EC 2012), and its physico-chemical and toxicological characteristics indicate that there may be a risk to aquatic environments due to releases of the chemical in Canada. A surveillance initiative across Canada has included sampling at 44 sites from July 2012 to March 2018. Triclosan was detected in 226 out of 918 samples, with concentrations ranging from less than 6 ng L^{-1} to 874 ng L^{-1}, and the detections averaging 54.23 ng L^{-1} (standard deviation; 97.6 ng L^{-1}). However, using the entire dataset (including censored data estimated with the Kaplan Meier model) the mean triclosan concentration was 17.95 ng L^{-1} and the standard deviation was 52.84 ng L^{-1}. Three samples at Wascana Creek (downstream), Saskatchewan, had concentrations above the Federal Environmental Quality Guidelines (FEQGs) of 470 ng L^{-1}, indicating a potential risk to the aquatic ecosystem. In this study, triclosan in samples collected downstream from municipal wastewater treatment plant discharges usually demonstrated higher concentrations than

upstream samples. Based on the results of this study, it is hypothesized that triclosan concentration have fluctuated between years of this study but not in an overall or significant increase or decreasing trend. Triclosan concentrations and detections are also more prevalent in urban than in rural or mixed development rivers. Performance evaluation of triclosan concentrations in the Canadian environment is scheduled to be re-assessed by 2024. Therefore a three year sampling program should be in place across Canada by 2021.

Keywords

Triclosan, monitoring, surface waters, trends, distributions

1.0 Introduction

Triclosan (5-chloro-2-(2,4-dichlorophenoxy)phenol) (CAS#3380-34-5) (Figure 1) is used as a material preservative and as an antimicrobial agent in a variety of consumer products to stop the growth of bacteria, fungi, mildew, and to deodorize (ECCC and HC 2016). It is widely used in personal care products (skin creams, toothpastes, soaps, deodorants, and body spray) (Halden 2014). There are no known natural sources of triclosan to the environment; its presence is due solely to human activities (ECCC and HC 2016). In 2001 triclosan was added to the list of priority chemicals for Screening Level Risk Assessment under the Canadian Environmental Protection Act (CEPA).

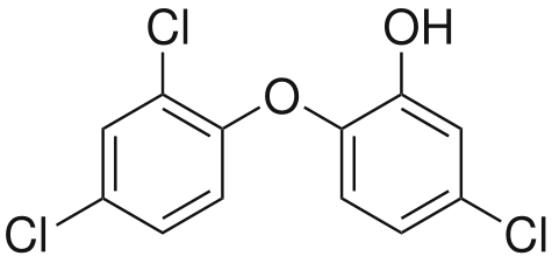

Figure. 1 Molecular structure of triclosan

Due to its low volatility, triclosan is not expected to partition to air (Halden 2014). Furthermore, owing to its use in many consumer products, which are for the most part released down the drain or sewers, triclosan reaches wastewater treatments plants (MWWTPs) where it is removed from wastewater at a high efficiency of 97-98% (Halden 2014). What is not removed is released to aquatic ecosystems as part of the MWWTP effluents (ECCC and

HC 2016). Industries that manufacture products containing triclosan may also release some into sewers (ECCC and HC 2016). Triclosan mostly partitions to water when discharged from MWWTP plants with a minor distribution to sediment (4.1 to 19.8% partition depending on pH) (ECCC and HC 2016). Since some triclosan partitions to sludge during the wastewater treatment process, it also reaches terrestrial ecosystems when sewage sludge is spread on land or deposited in landfills (Halden 2014 and ECCC and HC 2016).

Triclosan is not persistent in air, water, soil or sediment according to the *Persistence and Bioaccumulation Regulations* of the *Canadian Environmental Protection Act, 1999* (*CEPA* 1999) (ECCC and HC 2016), nor does it meet the bioaccumulation criterion (BCF or BAF≥5000) set out in these regulations (ECCC and HC 2016). Triclosan has a high inherent toxicity to a variety of aquatic organisms, such as algae, macrophyte, invertebrates, amphibians, and fish (ECCC and HC 2016). Continuous releases of triclosan in municipal wastewater and other sources may result in chronic exposure of aquatic organisms in downstream waters (ECCC and HC 2016). Endocrine disruption effects of triclosan have occurred in amphibians at environmentally relevant concentrations (Veldhoen et al 2006).

Concentrations of triclosan measured in Canadian sewage sludge have ranged from less than 0.1 to 46.4 ug g^{-1} and in surface water have ranged from below detection (0.10 ng L^{-1}) to 4160 ng L^{-1} (ECCC and HC 2016). The ECCC and HC (2016) risk assessment indicated that triclosan meets the criteria under paragraph 64(a) of CEPA as it is entering or may enter the environment in a quantity or concentration or under conditions that have or may have an immediate or long-term harmful effect on the environment or its biological diversity. ECCC has established a Federal Environmental Quality Guideline (FEQG) for triclosan to be 470 ng L^{-1} based on a chronic species sensitivity distribution for triclosan (ECCC 2017). The objectives of this project were to expand the network of surface water locations monitored, to assess the frequency of detection of triclosan in Canadian freshwaters, and to quantify triclosan concentrations.

2.0 Material and Methods

2.1 Monitoring Strategy

Triclosan concentrations are known to vary with population density (ECCC and HC 2016); therefore, sampling sites for this study were chosen to reflect a range of population densities across Canada. Attempts were also made to locate sites across Canada's major drainage areas where population densities would make it more likely for triclosan to be released into the environment (Figure 2). Furthermore, sampling sites were categorized in four groupings; reference, rural and mixed development, urban and MWWTP associated sites.

Two reference sites (Napan River, NB and upstream Mill Creek, BC) were chosen where upstream disturbance is minimal, while eleven sites were located in high-density urban sites (e.g., Still Creek, BC; Thames River, ON; Highland Creek, ON) (Table 1) where triclosan releases might be possible due to storm water and wastewater cross connections. Eleven sites were located in rural and/or mixed development areas which receives effluents from MWWTPs but the distance to the wastewater plants coupled with the high volume of river discharge at these sites make it unlikely that these sites would be highly impacted by triclosan (Table 1). Since MWWTPs have previously been found to be the most likely sources of triclosan (MacLeod and Wong 2010, Montagner et al 2014, Kimura et al 2014), nineteen sites were established immediately downstream of MWWTPs (Table 1). Of those nineteen sites, five were sampled both upstream (up) and downstream (down) of MWWTPs to assess the potential input of triclosan to the environment from a municipal wastewater source (Table 1). Those sites were located on the St John River, Grand River, Thames River, Red River and Wascana Creek (Table 1).

Quarterly or monthly sampling was conducted at 34 sites (Table 1) from July 2012 to March 2013. After March 2013, sampling frequency increased at sites where detections had occurred and decreased where detections were rare. Four sites located on the Red River, MB were added to the monitoring program in October 2013 and new sites in Ontario and Québec (Berthierville, QC, Princeville, QC; St Victor, QC, St Joseph de Beauce, QC; Trenton, ON; Prescott, ON; Credit River, ON, Beaverdam Creek, ON, Dicks Creek, ON) were added in 2015. This report includes triclosan data up to March 2017.

Characteristics of the sampling sites are listed in Table 1, including population (either in the whole watershed or only the largest urban centre upstream of each site), distance to the closest MWWTP, and river discharge (range or

annual average, if available). Watershed population was based on census data available on the web while discharge data (where available) are from ECCC hydrometric stations.

Table 1. List of monitored sites, sampling frequencies and geographical characteristics

Site Name, province	Sampling Frequency[a]	Latitude (DD)	Longitude (DD)	Watershed population	MWWTP (km)	Discharge (m^3/s)
Reference Sites						
Napan River, NB	Q	47.030	-65.3848	1 500	none	n/a
Mill Creek (reference), BC	Q	49.9835	-119.352	<50	none	n/a
Rural and Mixed Development Sites						
Saint John River (up)[b], NB	M, then Q	45.948	-66.869	< 5 000	36	350- 6 100
St. Lawrence River (Québec), QC	Q	46.806	-71.187	538 238	13	> 12 000
St. Lawrence River (Lavaltrie), QC	M	45.875	-73.281	3 824 221	29	> 10 000
Niagara River, ON	Q	43.255	-79.055	12 000 000+	15	5 796
St. Lawrence River (Wolfe) , ON	Q	44.211	-76.237	11 000 000+	90	6 850-9 200
Thames River (up) , ON	M	43.038	-81.223	< 40 000	33	5 - 70
Grand River (up) , ON	M	43.482	-80.481	15 000	30	n/a
Red River – Selkirk (up), MB	M	50.141	-96.869	633 450	28	>3200
Red River - Emerson, MB	Q	49.001	-97.223	665	4.5	>1 200
Wascana Creek (up), SK	M	50.416	-104.549	1 894	14	n/a
Osoyoos Lake, BC	Quarterly	49.004	-119.448	4 845	3	n/a
Okanagan River, BC	Quarterly	49.114	-119.566	4 824	8.75	5.9-85.5
Urban Sites						
Waterford River, NF	Q, then M	47.529	-52.741	24 284	none	1.1-3.3
Little Sackville River, NS	Q	44.763	-63.6888	21 379	none	0.017-8.52
Beaver Dams Creek, ON	various	43.103	-79.217	131 900	none	n/a
Dicks Creek, ON	various	43.154	-79.243	17 931	none	n/a
Mimico Creek, ON	M, then Q	43.646	-79.517	155 800	none	1 – 35
Taylor Creek, ON	M	43.701	-79.312	n/a	none	n/a
Highland Creek, ON	M	43.778	-79.191	360 000	none	1 - 40
Mill Creek (lower), BC	M, then Q	49.883	-119.499	117 312	none	19.4
Mill Creek (middle) , BC	M, then Q	49.887	-119.437	n/a	none	n/a
Still Creek, BC	M	49.259	-122.969	100 000	none	0.04-0.4
Serpentine River, BC	M, then Q	49.094	-122.801	n/a	none	6.23

Site Name, province	Sampling Frequency[a]	Latitude (DD)	Longitude (DD)	Watershed population	MWWTP (km)	Discharge (m^3/s)
MWWTP Associated Sites						
Saint John River (down)[c], NB	M, then Q	45.953	-66.624	56 224	0.25	500 - 7120
Berthierville, QC	B	46.087	-73.167	4091	0.25	n/a
Princeville, QC	B	46.205	-71.902	5693	0.25	n/a
St Joseph de Beauce, QC	B	46.336	-70.921	4722	0.25	12-1100
St Victor, QC	B	46.157	-70.919	2509	0.25	<5-60
St. Lawrence River (Trenton)	B	44.059	-77.334	11 000 000+	<1	n/a
St. Lawrence River (Prescott)	B	44.436	-75.285	11 000 000+	<1	n/a
Credit River, ON	various	43.909	-80.078	30729	0.75	0.75-10.25
Thames River (down) , ON	M	42.965	-81.389	366 151	6	10 - >500
Grand River (down) , ON	M	43.385	-80.385	507 096	5	25-300
Grand River @ Galt STP, ON	various	43.320	-80.314	>700 000	2	25-1100
Hamilton Harbour 914 , ON	M	43.268	-79.781	>500 000	1.5	n/a
Hamilton Harbour 909, ON	M	43.277	-79.878	>60 000	5	n/a
Hamilton Harbour 1001, ON	M	43.297	-79.802	>700 000	1.5	n/a
Hamilton Harbour 926, ON	M	43.183	-79.483	>175 000	0.25	n/a
Red River -Selkirk (down) , MB	Q	50.191	-96.844	9 934	2.75	>3200
Red River - Winnipeg, MB	Q	49.950	-97.098	633 450	0.1	n/a
Wascana Creek (down), SK	M	50.499	-104.800	193 100	8.5	<5 - 60
Okanagan River (North) , BC	Quarterly	49.479	-119.597	32 877	0.4	4.71-64.8

[a] Where B = bi-annually, M = monthly and Q = quarterly. [B] (up); upstream. [C] (down); downstream

2. Sample Collection

Most samples were collected by wading or by sampling pole. Samples from Hamilton Harbour, Berthierville and Osoyoos Lake were collected from a boat; others were also obtained from mid-span of bridges. Samplers avoided using triclosan-based hand cleansers before sampling, and used clean polyethylene gloves during the sampling procedure. All samples were collected in 1L contaminant free (trace clean) amber glass bottles, which were kept on ice and delivered to the analytical laboratory within 24 – 48 hours. Field measurements of temperature, conductivity, total dissolved solids (TDS), dissolved oxygen, pH and turbidity were made by water quality sonde. From March 2013 to February 2017, quality assurance and control samples, comprised of one blank and one

triplicate collection, were taken at seven sites. The field blank consisted of a 1L glass bottle filled with ultra-pure water (using MilliporeSigma water purification systems), which was exposed on site. Triplicate samples were collected using a single 4L amber glass bottle, which was mixed gently and sub-sampled, into three 1L bottles.

3. Laboratory Analysis

Laboratory analyses up to March 2014 were conducted by AXYS Analytical Services Ltd. in Sidney, BC, according to EPA method 1694 (LC/MS-MS) (EPA 2007) (AXYS 2014). Laboratory analysis for triclosan samples collected after March 2014 were done at Environment and Climate Change Canada's (ECCC) National Laboratory for Environmental Testing in Burlington, ON. Analysis was conducted according to laboratory method (B0779W), which is a direct inject LC-MS/MS analysis. 950 uL of sample is spiked with 50 uL of an internal standard mix, containing the surrogate compounds - $^{13}C_{12}$-triclosan. The sample is separated using a Waters BEH C18 UPLC column (100 x 2.1 mm, 1.7 um particle size) and analyzed using a Waters TQ-S LC-MS/MS system. A minimum of one ion transition is monitored for each analyte, two where possible (J Small, ECCC, pers. Comm., November 2017). Acquisitions were done by MRM mode while ionization was performed by negative ion electrospray at both laboratories. The detection limit for the AXYS methodology averaged 6.02 ng L^{-1} while the detection limit for Environment Canada's methodology was 17.7 ng L^{-1}. The LOQ for triclosan analysis for the AXYS methodology was 5 ng/L. Initial calibration of the LC-MS/MS instrument was performed by the analysis of five or more calibration solutions (AXYS 2014). A mid-level calibration is also analysed after every 20th sample (AXYS 2014). A calculated concentration (0.093 ng/mL) and standard deviation (0.0049 ng/mL) for triclosan based on ten spiked Lake Ontario water samples (spike of 0.1 ng/mL) was performed by the ECCC laboratory.

The analytical laboratories of NLET and AXYS are accredited by CALA (the Canadian association for laboratory accreditation) to the standard ISO/IEC 17025. NLET is an active participant in major national and international inter-laboratory performance-testing programs. AXYS also holds analysis accreditation with the NELAC institute.

2.4 Statistical Treatment

All statistical analysis were conducted using Systat™11, Excel™2010 and R. Non-detect values were included using the Kaplan-Meier method (Helsel 2012) to calculate means and standard deviations. Censored box plots were created with the help of the NADA package in R (Helsel and Lee 2006). Censored data were also used in the estimations of differences in groups (non parametric Wilcoxon) or regressions by MLE with the help of the NADA package for R (Helsel and Lee 2006). The NADA package uses survival analysis methods to estimate descriptive statistics. For triplicate samples, and where all three samples had similar detections, the first sample was designated the discrete sample, while the other two results were assessed for data quality purposes only. Relationships between log transformed field measurements and log transformed triclosan concentrations were evaluated using simple linear regressions, as were temporal trends of triclosan concentrations.

3. Results and Discussion

1. Quality Control and Quality Assurance

For all the measurements obtained from AXYS, one laboratory blank and one spiked matrix sample were analysed with each batch of samples. The recovery rate for 13-C triclosan for all samples averaged 62.9% and laboratory blanks were below method detection limits for all but five out of the 41 samples (detections were 5.31, 6.65, 7.01, 12.9 and 22.7 ng L^{-1}). All data presented in this report are deemed acceptable against the analytical laboratory's QC specification for blanks and matrix spike (AXYS 2014). Blanks and spiked matrix were performed on batches of samples at the Environment Canada laboratory. The number of samples per batches varied from 5 to 10 sampling sites. Overall, 44 blank samples were analysed and all returned values of non detect (<17.7 ng L^{-1}). Only six out of the 52 spiked matrix samples were outside the acceptable recoveries of between 70 and 130%. The values obtained from these batches of samples were carefully examined to ensure their validity.

2. Summary of Water Quality and Triclosan Results

A summary of field water quality measurements is provided in Table 2, demonstrating the variety of conditions across which measurements were made. The measurements in Table 2 include all available measurements for the first three years of the monitoring program.

Table 2. Summary of in-situ water quality measurements at triclosan sampling sites across Canada.

Parameter	Range	Mean	Standard deviation
Temperature (°C)	-0.24-25.4	9.93	7.11
Conductivity (us cm^{-1})	0.5-1945	322.3	451.2
Total Dissolved Solids (TDS) (mg L^{-1})	0.29-8.6	1.21	1.80
Dissolved oxygen (mg L^{-1})	1.58-16.49	11.14	7.9
pH	6.14-9.04	7.91	0.49
Turbidity (NTU)	0.38-129	10.5	19.3

Linear regressions between field measurements and triclosan concentrations revealed weak but significant relationships for triclosan with total dissolved solids (TDS) and pH: $\log_{10}(\text{triclosan}) = 1.284+0.412*\log_{10}\text{TDS}$ (n=39, R^2=0.13, p=0.027) and $\log_{10}(\text{triclosan}) = 2.998-0.227*\text{pH}$ (n=78, R^2=0.072, p=0.017). A TDS measurement represents the total of organic and inorganic particles small enough to pass through a 2 um pore size filter. The positive relationship with TDS may be due to the moderately sorptive characteristic of triclosan (Koc up to 4.67) (HC and EC 2012).

A summary of triclosan detections and concentrations are presented in Table 3 and Figure 2. Triclosan has been detected in 226 out of 918 samples, with concentrations ranging from 5.1 to 874 ng L^{-1}, and a mean of 54.23 ng L^{-1} for the samples, which had detections. A Kaplan-Meier model using the censored data reveal a mean of 17.95ng L^{-1}

. In comparison, ECCC and HC (2016) published a range of Canadian concentrations of triclosan in surface water from less than MDL to 4160 ng L^{-1}. Triclosan was not detected in any of the samples at six sites sampled frequently in this study (Table 3). Triclosan was detected at a frequency of 1-24% at twenty five sites, between 25-49% at eight sites, and greater than 50% at five sites (Table 3). The mean and standard deviations below in Table 3 were calculated using the Kaplan-Meier model for censored data (Helsel 2012). The sampling sites with the highest triclosan concentrations include (in decreasing order), Wascana Creek (downstream), Grand River, Hamilton Harbour, Berthierville and Taylor Creek (Table 3).

Table 3. Summary of triclosan detection frequencies and concentrations at 44 sites in Canada

Site name	No. detections / No. samples	Minimum (ng L^{-1})	Maximum (ng L^{-1})	Median (ng L^{-1})	Mean* (ng L^{-1})	Standard deviation* (ng L^{-1})
Non Urban/ Reference Sites						
Napan River	0/7					
Mill Creek (reference)	4/18	7.8	35.3	20.6	5.65	12.77
Rural and Mixed Development Sites						
Saint John River (upstream)	3/23	5.7	8.0	6.4	1.44	4.36
St. Lawrence River (Québec)	3/20	6.9	151	7.7	8.9	40.2
St. Lawrence River (Lavaltrie)	11/43	6.1	61.5	8.4	8.21	12.67
Niagara River	2/15	7.1	7.5	7.3	2.92	8.78
St. Lawrence River (Wolfe Island)	1/11	74.6	74.6	74.6	6.21	
Thames River (upstream)	3/37	14.3	111.0	19.1	4.42	22.99
Grand River (upstream)	2/37	5.2	6.7	5.95	0.85	4.81
Red River – Selkirk	2/28	9.8	14.0	11.9	11.88	10.6
Red River – Emerson	0/10					
Wascana Creek (upstream)	2/35	64.5	143	103.8	5.93	36.55
Osoyoos Lake	1/10	42.0	42.0	42.0	3.82	
Okanagan River	1/7	8.9	8.9	8.9	1.49	
Urban Sites						
Waterford River	6/25	6.4	17.0	9.4	5.44	9.14

Site name	No. detections / No. samples	Minimum (ng L^{-1})	Maximum (ng L^{-1})	Median (ng L^{-1})	Mean* (ng L^{-1})	Standard deviation* (ng L^{-1})
Little Sackville River	5/14	5.0	25.4	8.5	9.18	7.29
Beaver Dams Creek	1/20	77.7	77.7	77.7	3.88	
Dicks Creek	3/21	5.8	156.0	62.6	15.69	41.17
Mimico Creek	3/20	5.6	80.4	23.6	6.10	22.22
Taylor Creek	19/35	11.5	119.0	23.4	23.01	24.67
Highland Creek	11/36	5.4	142.0	11.8	10.30	25.34
Mill Creek (Lower)	3/23	5.86	111	7.6	5.86	27.74
Mill Creek (Middle)	4/16	11.6	20.7	17.1	6.10	22.22
Still Creek	11/41	5.20	162.0	17.7	10.78	29.58
Serpentine River	3/17	7.9	11.3	7.9	2.04	5.31
MWWTPs Associated Sites						
Saint John River (downstream)	2/24	6.0	6.0	6.0	0.85	3.8
Berthierville	2/8	30.9	177.0	104.0	25.99	81.98
Princeville	1/5	20.7	20.7	20.7	4.14	
St Joseph de Beauce	0/6					
St Victor	0/6					
St. Lawrence River (Trenton)	0/4					
St. Lawrence River (Prescott)	0/4					
Credit River	1/6	60.1	60.1	60.1	10.02	
Thames River (downstream)	13/37	7.5	193.0	11	14.97	35.10
Grand River(downstream)	26/40	5.9	137.0	22.1	26.91	33.48
Grand River @ Galt STP	3/6	20.4	269.0	46.2	55.93	118.47
Hamilton Harbour 914	26/44	8.9	268.0	57.6	46.44	64.23
Hamilton Harbour 909	2/31	6.0	6.1	6.0	1.21	6.02
Hamilton Harbour 1001	11/32	5.5	122.0	6.3	10.64	23.39
Hamilton Harbour 926	7/31	7.9	105.0	15.2	8.06	21.42
Red River – Selkirk (downstream)	1/10	5.7	5.7	5.7	5.73	
Red River - Winnipeg	2/10	19.3	37.1	28.2	5.64	16.92

Site name	No. detections / No. samples	Minimum (ng L^{-1})	Maximum (ng L^{-1})	Median (ng L^{-1})	Mean* (ng L^{-1})	Standard deviation* (ng L^{-1})
Wascana Creek (downstream)	23/37	13.8	874.0	144.0	136.93	196.43
Okanagan River (North)	3/8	7.4	17.0	12.6	5.28	8.67

*Mean and standard deviations were calculated using the Kaplan Meier method (Helsel 2012)

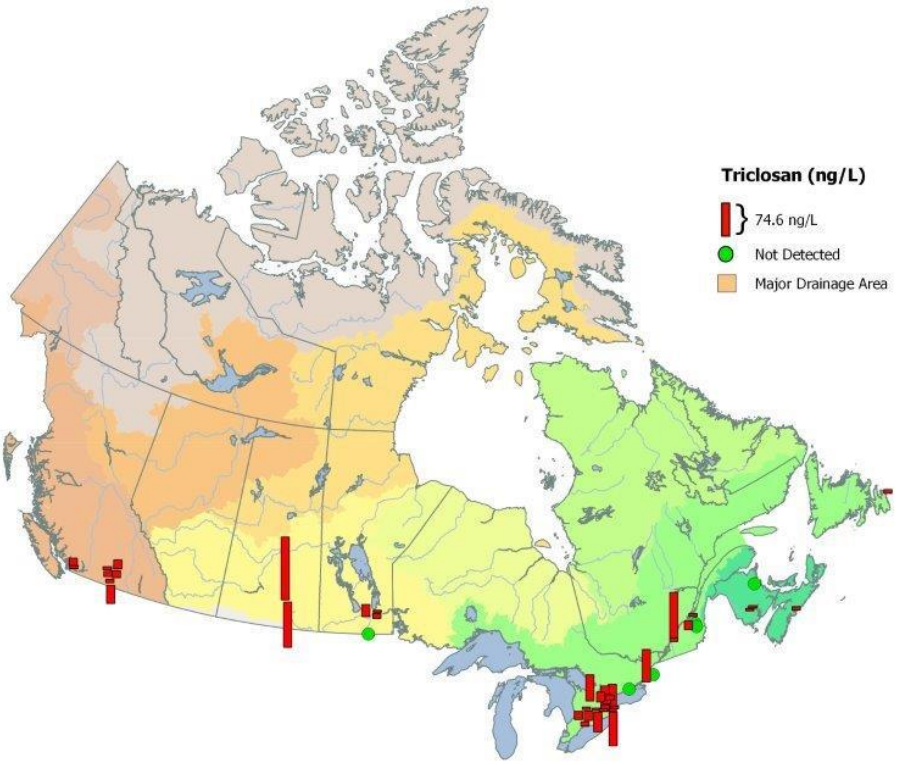

Figure. 2 Median triclosan concentrations in Canadian major drainage areas.

In our study, the FEQG was exceeded at one sampling site (Wascana downstream) on three occasions (572 to 874 ng L^{-1}), indicating a potential ecological risk. Elevated but lower than FEQG values were also collected at Wascana downstream (111 to 395 ng L^{-1}), Grand River @Galt STP (269 ng L^{-1}), Hamilton Harbour 914 (191 to 268 ng L^{-1}),

downstream Thames River, (193 ng L^{-1}), Berthierville, (177 ng L^{-1}), Dicks Creek (156 ng L^{-1}) and Still Creek, BC (162 ng L^{-1}).

Although triclosan is generally thought to enter the aquatic environment primarily from wastewater treatment plants, thirteen sites (in three watersheds) in our study were not directly associated with MWWTP discharges to provide baseline spatial triclosan concentrations on a national scale. A description of each sampling site and a summary of triclosan concentrations at each site are provided in the following paragraphs. Where data were available, dates of detections and maximum concentrations were compared with rainfall and river discharge data at the time of collection. No statistically significant relationships between triclosan concentration and river flow or rainfall were found at any of these sites.

3.3 Reference Sampling Sites

The Napan River is a tributary to the Miramichi River and it drains a rural/farming area with a former air force base in its headwaters. There were no known direct sources of triclosan of the river, and there was no detections of triclosan at this site on the seven sampling occasions. Mill Creek is located in the central Okanagan Valley. Its headwaters are located in the surrounding mountains of the Interior Plateau and it flows for 33 kms until it joins Lake Okanagan in the City of Kelowna. The Mill Creek reference site is well upstream of Kelowna. Using the Kaplan Meier method, the mean concentration of triclosan was 5.65 ng L^{-1} (22% detections) at the reference site. There are no known direct sources of triclosan upstream of the Mill Creek reference site however there is limited anthropogenic activity including: cottages 11km upstream, active logging, free-range cattle ranching, off-road vehicles and summer time bathing. Since the mean value is very close to the detection limit of the laboratory, overall the concentrations of triclosan at this site is reflective of a non-impacted site. Section 3.1 details the issues concerning the detection at this reference site.

3.4 Rural or Mixed Development Sampling Sites

The Québec and Lavaltrie sites are both on the St. Lawrence River, downstream of MWWTPs in Quebec (13 km) and Montreal (10 km), respectively. The detection frequency at Lavaltrie were higher than the Québec City site (26% compared to 15%) (Table 3), which is probably a function of a similar size receiving environment with a much greater population and associated wastewater volumes in Montréal than Quebec City. Despite a higher detection frequency in Montreal, concentrations in Quebec overall were greater, with a maximum value of 151 ng L-1 at the Québec site in January 2015. The mean concentrations at both these sites are very similar with values of 8.9 and 8.21 ng L-1 at Quebec and Lavaltrie respectively (Table 3) The Niagara River site is located downstream of Lake Erie while the St Lawrence River site (Wolf Island) is located 15 km downstream of Lake Ontario. Triclosan was detected in only two out of fifteen samples at Niagara River (7.1 and 7.5 ng L^{-1}) and only detected once (74.6 ng L^{-1}) in eleven samples at Wolfe Island. Both sites encompass large populations in their upstream watersheds, though the Wolf Island Site has no large centres within 50 km. The high frequency of non-detects is likely due to the high dilution of the Great Lakes watershed and to the moderate size of the closest MWWTPs to those sites. The Red River site at Emerson is located on the border between the United States and the province of Manitoba. The largest urban centre upstream of Emerson is Grand Forks (North Dakota), located 120 km upstream with a population of 52 838 citizens. Triclosan was not detected in any of the ten samples collected to date at this site. Concentrations of reference samples at Osoyoos Lake were collected near the middle of the lake and there was one triclosan detection (42 ng L^{-1}) in the ten samples collected at that site. Overall, the mean concentration at this site (3.82 ng L^{-1}) is reflective of a reference site as well. The Okanagan River site encompasses a mix of vineyards, fruit farms, livestock, and some residential areas. There has only been one detection out of seven samples of triclosan at the Okanagan River site (8.91 ng L^{-1}), which was not associated with a significant rainfall or rise in water level. The source of that detection could have been the multiple small or medium MWWTPs located further upstream.

3.5 Urban Sampling Sites

Both the Waterford and the Little Sackville Rivers are downstream of urbanized and residential areas. Triclosan was detected at both sites in approximately one third of samples with means of 5.44 ng $L^{-1}$and 9.18 ng L^{-1}at Waterford and Little Sackville, respectively (Table 3). Mimico Creek, Taylor Creek and Highland Creek are all urban watercourses in or near Toronto, with >85% urbanization upstream. All sites had frequent and/or higher detections of triclosan in samples collected with maximum concentrations of 80.4 ng L^{-1}, 119 ng L^{-1} and 142 ng L^{-1} at Mimico, Taylor and Highland, respectively (Table 3). Triclosan in these waters is likely due to storm sewer outfalls, combine sewer-stormwater outfalls, cross connections and breaks in sanitary lines which are often associated with pervasive urbanization. Another site was added in 2015 on the Beaver Dams Creek near St Catherines, ON. This particular location was chosen as it receives effluent from two paper recycling mills located upstream. Only one detection of triclosan occurred on this creek (26.3ng L^{-1}) while nineteen samples were non detects. The presence of the paper recycling mills may be the cause of the detection as triclosan was sometimes applied to paper products (Halden 2014). Finally, a last site was added in 2015 on Dicks Creek. Dicks Creek is a tributary of the Twelve Mile Creek and flows through residential, industrial and commercial areas. Three out of twenty one samples had detections of triclosan (5.84 to156ng L^{-1}, mean = 15.69 ng L^{-1}). There is no MWWTP located upstream of this location, therefore triclosan detections may be due to cross connections in storm water and sewage lines or from the general urbanization of this stream.

Sampling also occurred on two more sites on Mill Creek, BC. One site is located in the middle of the city (Mill Creek Middle) with mostly industrial areas upstream, while the last one (Mill Creek Lower) is located at the mouth of Mill Creek, just before it flows into Lake Okanagan. Mill Creek averaged 6.10 and 5.86 ng L^{-1} (with 25% and 13% detections) at the middle and lower sites, respectively. Both of the middle and lower sites have commercial and residential areas located upstream. Flow from storm sewers might discharge into Mill creek near those sites, possibly contributing triclosan from cross connections or cracks in the sanitary lines. Of particular note is the detection of 111 ng L^{-1} at one occasion at the Mill Creek (Lower) site. The mean triclosan concentrations at both Mill Creek sites are very similar (Table 3). The censored boxplots (Figure 3) presents the triclosan concentrations for all three sites on Mill Creek for comparison purposes. Most of the data in Figure 3 is under the later detection limit (17ng L-1). The highest range and overall concentrations were detected at the Mill Creek Lower site (Figure 3). Using the cendiff command in R reveals that the distribution between groups is not statistically different ($p=0.275$).

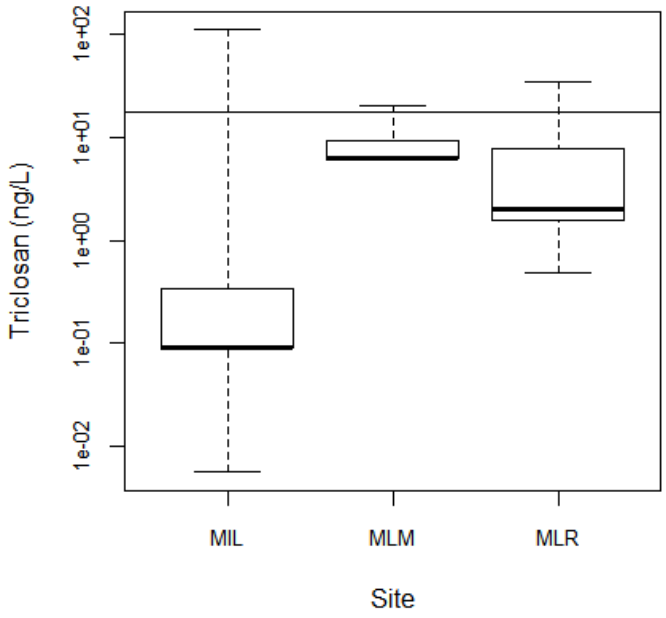

Figure 3. Censored Boxplots of Mill Creek triclosan concentrations as a function of location (lower (MIL), middle (MLM) and reference (MLR)). The horizontal line in the box plot represents the detection limit for triclosan concentrations.

The Serpentine River (Surrey, BC) watershed area is 154.2 km^2, with heavy urbanization in its upper reaches and agriculture and channelization dominating the downstream area. The Serpentine River only has an 18% detection rate of triclosan. Environment and Climate Change Canada does not monitor the discharge of Serpentine Creek, however a nearby river had a large increase in discharge a week before the highest concentration measured (11.3 ng L^{-1}). Therefore, the elevated triclosan concentration may have been due to increased urban storm sewer discharge into the Creek. Still Creek is located in Burnaby, BC, which is dominated by industrial and residential areas upstream. The upper reaches of the watershed are enclosed in storm sewers or culverts and only 3.25 km remains open (Lees and Associates et al 2002). The creek is subject to water runoff during heavy rain and there are documented cross connections of storm water and municipal wastewater lines (Lees and Associates et al 2002). The

watershed's high imperviousness reduces the opportunity for infiltration of runoff from residential, industrial and roadways, therefore triclosan detections are more likely due to leaks in those storm water and sanitary cross connections. Concentrations of 162 and 79.6 ng L^{-1}were also detected on Still Creek in July and September 2015. The September detection coincided with a large rainfall event where the creek level increased by about 90 cm.

3.6 MWWTPs Associated Sites

Five rivers and were monitored both upstream and downstream of municipal wastewater treatment plants (Saint John River, Thames River, Grand River, Wascana Creek, Red River) to evaluate the contribution of those potential sources of triclosan to the aquatic receiving environment. In addition, four sites in Quebec (Berthierville, Princeville, St Victor and St Joseph de Beauce) and two in Ontario (Prescott and Trenton) were located immediately downstream of MWWTP which receive effluent from textile plants. The sites on the Credit River (ON) and Okanagan River (North) were located downstream of MWWTP but no upstream sampled were collected at these locations. Sites in Hamilton Harbour were also located close to the different MWWTP outfalls on purpose.

The sampling sites on the Saint John River are approximately 20 km apart. The upstream site lies upstream of the Mactaquac dam, with several small towns and associated MWWTPs upstream (Table 1). The downstream site is just below the plume of the Fredericton MWWTP. Average concentrations of triclosan upstream and downstream of Fredericton on the St. John River were very similar (1.44 and 0.85 ng L^{-1} respectively) (Table 3), suggesting that triclosan in the St. John River is not closely associated with its largest urban centre, but seems to be ubiquitous in the system from smaller points of discharges throughout its watershed.

In 2015, four additional sites were sampled in the province of Québec; St Joseph de Beauce, Berthierville, Princeville and St Victor. All four sampling sites were chosen as they were located downstream of the effluent from a municipal wastewater treatment plants which received effluents from textile factories. Two detections of triclosan occurred in Berthierville (30.9 and 177ng L^{-1}) on a branch of the St Lawrence River. The elevated concentration

detected at this location is significant since the MWWTP effluent (mean of 0.086 m^3/s) gets highly diluted at its discharge. The stream which receives the MWWTP effluent from Princeville is very small (only 1.5m across) and 1 out of five samples had a triclosan detection (20.7 ng L^{-1}). The MWWTP for St Joseph de Beauce is the Chaudière River which has a maximum discharge volume of $1200m^3/sec$ while the MWWTP discharge is approximately $0.067m^3/s$ (Quebec 2013). Finally, the MWWTP discharge from St Victor is approximately $0.042m^3/s$ while the maximum discharge of the Bras Victor can attain $60m^3/s$. Both rivers (St Victor and St Joseph de Beauce) have the potential to dilute the triclosan concentrations to a non-detect level considering the low volume of MWWTP discharge compared to the volume of water in the rivers, which would explain the lack of detections at these two sites. The sampling sites in Prescott and Trenton both had no detections in the four samples collected. Although both of these sites were located close to the discharge of a MWWTP, the discharge occurs in the St Lawrence River, which is probably diluting any triclosan present to a non-detectable concentration.

The upstream Thames River site had detections of triclosan in only 8% of samples (but an average of 4.42 ng L^{-1}) while the downstream site had 35% detections and an average of 14.97 ng L^{-1} (Table 3). The frequency of detections and mean results were intuitive given the large influence of the city of London and its MWWTPs at the downstream site as well as possible contributions from the main branch of the Thames River, from the MWWTPs at Ingersoll and Woodstock. However, the smaller communities upstream of London (e.g. St. Mary's and Stratford) may sometimes contribute a significant concentration of triclosan to the Thames River as there was a detection of triclosan of 111 ng L^{-1} on one sampling date at the Thames upstream station (Table 3). The censored box plot of triclosan concentrations on the Thames River clearly indicates a contribution of London's MWWTP (Figure 4). Both the spread and median of the data in the boxplots are higher in the downstream site (Figure 4), and the two group test (cendiff command in R) reveals a significant difference ($p=0.0415$).

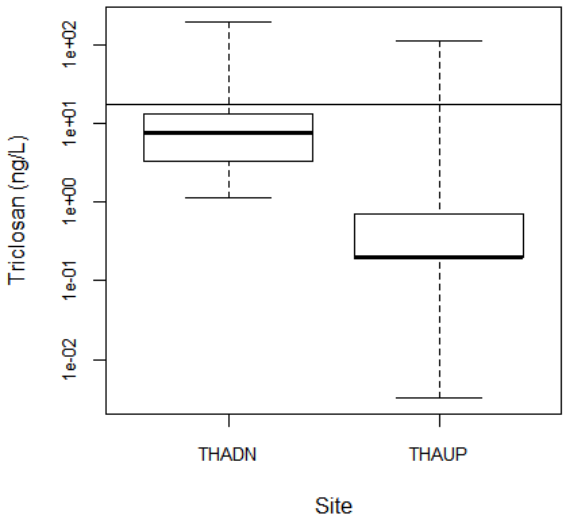

Figure 4. Censored Boxplots of Thames River (THA) triclosan concentrations as a function of location (upstream(UP) and downstream (DN) of the MWWTP)

The differences in triclosan concentrations between Kitchener-Waterloo upstream and downstream sites were very noticeable in the Grand River (Table 3). The mean concentration and frequency of detections were higher at the downstream site (26.91 ng L^{-1} and 65%) compared with the upstream site (0.85 ng L^{-1}, 5%) and did not correlate with daily discharge of the Grand River. The smaller detection rate and mean concentrations upstream probably reflects the smaller but important contribution of triclosan to the Grand River from the smaller urban centres located upstream of Kitchener-Waterloo. Figure 5 also indicate a larger spread and median of triclosan concentrations for the downstream site indicating the MWWTP as a potential source of triclosan. The two groups test (cendiff command in R) reveals a statistically significant difference between the two groups ($p<0.001$).

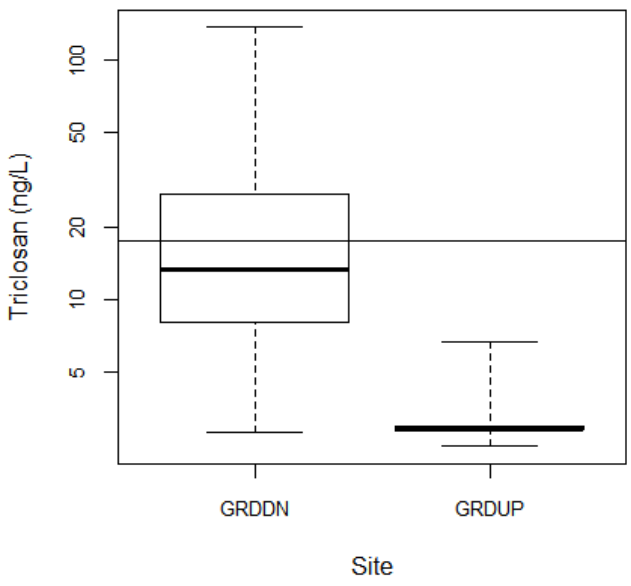

Figure 5. Censored Boxplots of Grand River (GRD) triclosan concentrations as a function of location (upstream (UP) and downstream (DN) of the MWWTP)

In 2015 an additional sampling location was added on the Grand River downstream of the Galt MWWTP, which services the town of Cambridge (population of 126 748). Of the six samples collected from that site, three were non detect while the others measured between 20.4 to 269 ng L^{-1} (mean was 55.93 ng L^{-1}). The triclosan detection may be explained by the presence of triclosan in the effluent from the Galt MWWTP as well as the presence of triclosan in the effluent from the larger Kitchener-Waterloo MWWTPs located upstream of this station. In 2015, a site was added to this project and it was located on the Credit River near Orangeville, ON which as a population of 30 729. Of the first six samples collected, only one sample contained triclosan (60.1 ng L^{-1}, Table 3). This site is located in the upper reaches of the Credit River, downstream of the Orangeville STP and below the Orangeville Reservoir. The lack of triclosan detection on 5 out of 6 samples is surprising due to the close proximity of the MWWTP outfall.

Hamilton Harbour is a 2150-hectare bay at the western tip of Lake Ontario. There were four sampling sites located in Hamilton Harbour. HH 1001 is located in the middle of the harbour while the three others are close to three MWWTP discharges in the southeast corner (HH 914), west end (HH 909) and northeast corner (HH 926) of Hamilton Harbour. There was a 34% rate of detection of triclosan at HH1001, a site that was strategically located to encompass all sources of discharge of Hamilton Harbour before entering Lake Ontario. HH 914, located near the Hamilton MWWTP, had the most frequent detections (26/44) and highest concentration and mean of triclosan (268 and 46.44 ng L^{-1}respectively) in Hamilton Harbour, which also had the fifth highest concentration of triclosan in this study (Table 3). HH 909, located close to the Dundas and Waterdown MWWTP, had detections 6% of the time and the lowest mean concentration (1.21 ng L^{-1}) of all the sites in Hamilton Harbour. HH 926, located close to the outfall of the Burlington, MWWTP, had detections in 23% of the samples and the second highest mean concentration in the Harbour (8.06 ng L^{-1})(Table 3). Triclosan concentrations in Hamilton Harbour sites did not fluctuate at the same rate, and higher concentrations did not coincide with any large rainfall events. Overall, triclosan concentrations in Hamilton Harbour appear to be related to the proximity and operating size of the MWWTPs, although triclosan concentrations seem to be pervasive in the Harbour as the sampling point closest to the discharge into Lake Ontario (HH1001) has a mean concentration of 10.64 ng L^{-1} (Table 3).

There are four sites located on the Red River, which has a maximum daily discharge of above 3200 $m^3 s^{-1}$ and highest flows in May. One is at Emerson, just north of the International boundary and well upstream of the influence of Winnipeg. Results from that site were discussed in the reference section. The second site (Red River – Winnipeg) is located immediately downstream of Winnipeg's North End MWWTP and ultimately receives discharges from the other two Winnipeg MWWTPs as well. The third site is located downstream of Winnipeg in the City of Selkirk and the fourth is located 2.75 km downstream of Selkirk's MWWTP. Triclosan was detected on two occasions at the Winnipeg and once at the Selkirk MWWTP sites with average concentrations of 5.64 and 5.73 ng L^{-1} respectively. A small precipitation event of 6.8 mm occurred on the day prior to the 37.1 ng L^{-1} detection (Winnipeg site) and may have been a contributing factor, though such a small rain is unlikely to have triggered a MWWTP overflow event. Again results seem to suggest a positive relationship between the size of the MWWTP

and the concentration of triclosan detected. The site located upstream of Selkirk had a 10% detection rate and averaged 11.88 ng L^{-1} which seems to suggest some influence of Winnipeg's MWWTPs.

Wascana Creek, in Regina, Saskatchewan, had the greatest upstream to downstream difference in triclosan concentrations of any other in this study (Figure 6). The mean triclosan concentration downstream was 136.93 ng L^{-1} (65% detections) and ranged from 13.8 to 874 ng L^{-1}, while the upstream site had a 6% detection rate of triclosan and a mean of 5.93ng L^{-1} (Table 3). The censored boxplot indicates a large difference in median and data spread of the two sites with the downstream site noticeably higher than the upstream site. As expected, the two groups test (cendiff command in R) revealed a statistically significant difference between the two groups ($p<0.001$). The only three detections above the FEQG value occurred at this site in various years (2014 and twice in 2016). The results indicate that the small communities located upstream on Wascana Creek do not contribute significantly to the triclosan concentrations measured downstream of Regina, and that downstream concentrations are largely due to the discharge from Regina's MWWTP. Waiser et al. (2011) measured triclosan up to 110 ng L^{-1} 10 km downstream of Regina's MWWTP. They concluded that the elevated pharmaceuticals in Wascana Creek were due to the creek's hydrograph, with treated sewage effluent making up almost 100% of the flow from October to March and only had a maximum dilution of 9.4 times over the spring and summer (Waiser et al. 2011). Our maximum values which occurred in January and December seem to corroborate the results from Waiser et al. (2011).

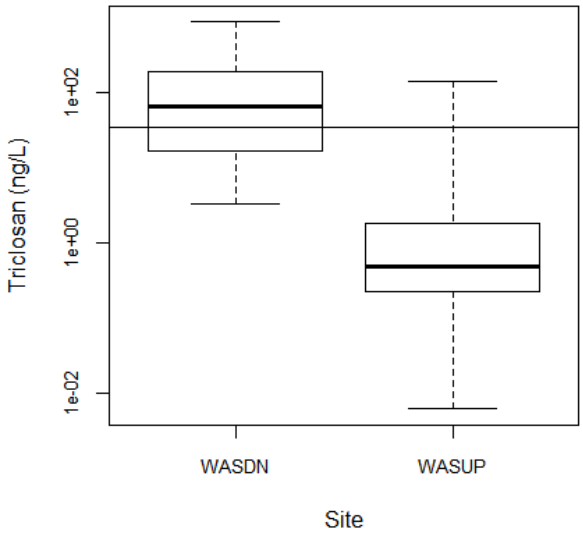

Figure 6. Censored Boxplots of Wascana Creek triclosan concentrations as a function of location (upstream (Wasup) and downstream (Wasdn) of the MWWTP)

The Okanagan River North site is located about 150 m downstream of the Penticton MWWTP outfall, and is adjacent to commercial and residential areas. There is no upstream sampling site for this location. There were detections of triclosan in 38% of the samples from that site, at concentrations between 7.4 and 17 ng L^{-1} with a mean of 5.28 ng L^{-1} (Table 3). The low rate of detections might be related to the relatively small population (32 877 citizens, Table 1) serviced by the MWWTP as well as the dilution of the Okanagan River. The Penticton MWWTP releases approximately 0.15 m^3/s to the Okanagan River, which at that location has a discharge volume of approximately 5-65 m^3/s.

3. Temporal Trends

A boxplot of the censored data of all sites combined as a function of year reveals similar spreads of concentrations for all years (Figure 7). A non-parametric Wilcoxon test (cendiff command in R, Helsel and Lee 2006) reveals a significant difference between groups (years) ($p<0.001$). However, a subsequent censored regression by year yielded a non-significant p value ($p=0.205$). Therefore it is hypothesized that triclosan concentration in this study have fluctuated between years of this study but not in an overall increase or decreasing trend. The difference in triclosan concentrations between years may be partially explained by the change in laboratory and associated increase in detection limit which occurred in 2015 as well as the addition or deletion of sampling sites between years. Interestingly, the highest mean was for the 2017 samples while the lowest was for the 2012 samples (71.64 and 10.4 ng L^{-1}).

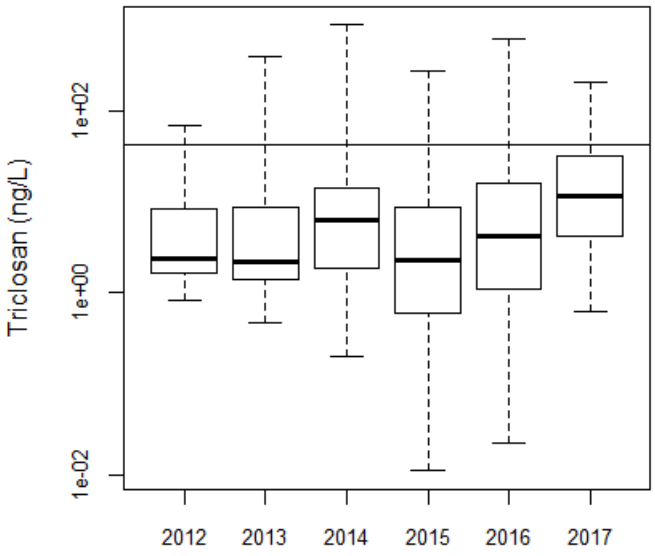

Figure 7. Censured box plots of censored triclosan concentrations as a function of year of sampling

Samples of triclosan were collected in all months of the year throughout Canada. A season category was created to analyse the changes (if any) of triclosan based on the months of sampling. The season categories were as follows: 1 (January to March), 2 (April to June), 3 (July to September) and 4 (October to December) (Figure 5). A boxplot of the censored data by season reveals similar spreads of concentrations for all seasons (Figure 5). A subsequent non-parametric Wilcoxon (cendiff command in R) resulted in a non-significant difference between seasons. Although visually, the Wascana Creek (downstream) samples collected in winter drive the spread of the data higher for the winter season in Figure 8.

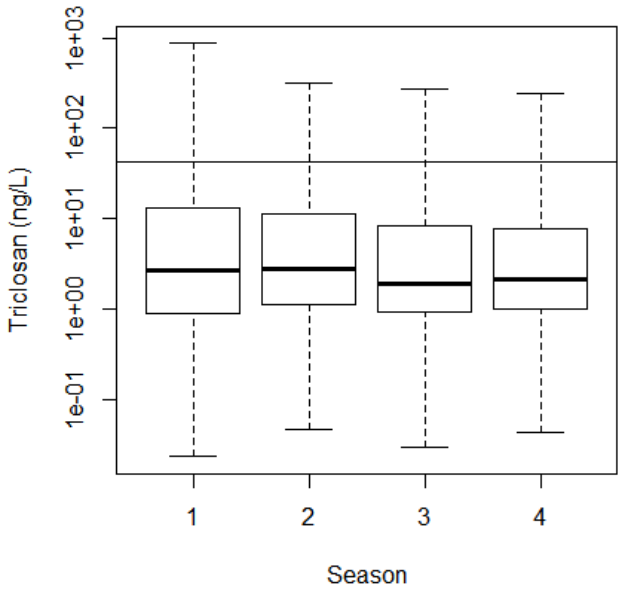

Figure 8. Box plots of censored triclosan concentrations as a function of seasons of sampling

Similarly, in a year-long study in Alberta (Canada), MacLeod and Wong (2010) did not detect any significant temporal variations in triclosan concentrations downstream of a MWWTP. To date there have been no changes to legislation in Canada related to triclosan manufacture, sale or use, so significant temporal changes were not expected. The evaluation of temporal patterns will be ongoing as more data are collected at these sites, particularly if legislative changes occur.

3.8 Spatial Trends

A non-parametric Wilcoxon test (cendiff command in R) (with ocean drainage areas as a category) reveals a significant differences between groups ($p<0.001$). The differences between groups is partially driven by the results from the sites located in the Hudson Bay Drainage Area (mean = 44.9 ng/ L^{-1}). However, the main difference in the triclosan concentrations between Hudson Bay and the rest of the drainage areas can be explained by the inclusion of Wascana Creek in the Hudson Bay watershed. Wascana Creek (downstream) had shown the two highest concentrations of this study and has repeatedly had significant triclosan detections due to the close proximity of the MWWTP and the low natural flow of this creek in the winter months. Figure 6 below indicates clearly the large span in concentrations occurring at the sites in the Hudson Bay watershed in comparison to all the other watersheds sampled in this study. Figure 10 clearly indicated the higher values of triclosan are largely for the Hudson Bay drainage.

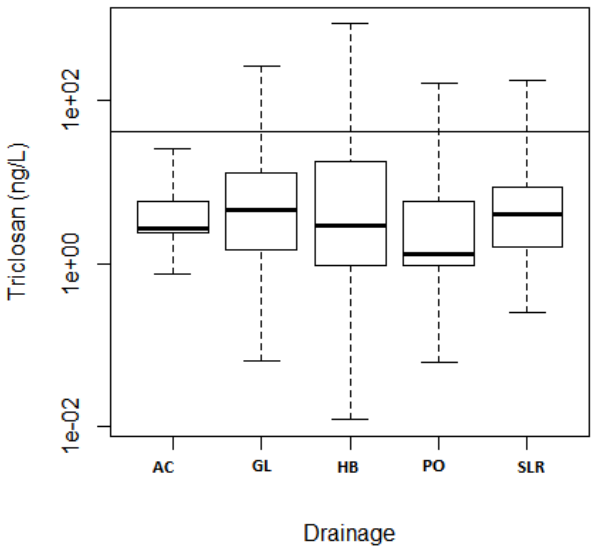

Figure 9. Box plots of censored triclosan concentrations as a function of drainage areas (whereas (Atlantic Canada (AC), St Laurent River (SLR), Great Lakes (GL), Hudson Bay (HB) and Pacific Ocean(PO))

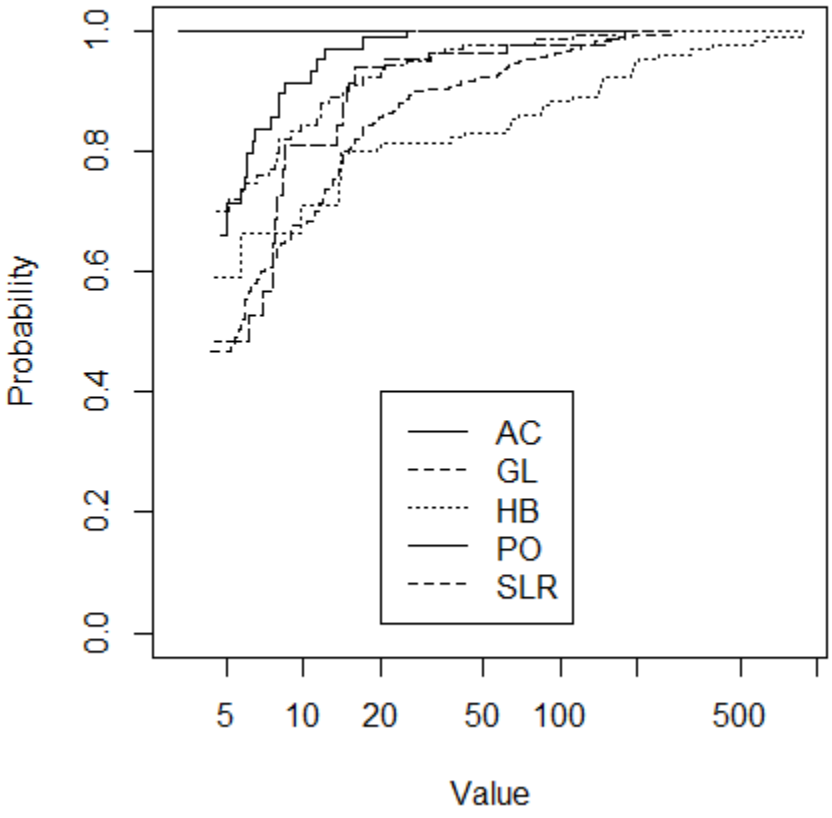

Figure 10. Triclosan concentrations per ocean drainage areas of Canada (whereas (Atlantic Canada (AC), St Laurent River (SLR), Great Lakes (GL), Hudson Bay (HB) and Pacific Ocean(PO)).

Lastly, a comparison was made of triclosan concentrations between the four groupings of rivers in this study; reference, mixed and rural sites, MWWTP sites and urban sites. A non-parametric Wilcoxon test (cendiff command in R) (with river groupings as a category) reveals a significant differences between groups ($p<0.001$). The censored boxplot (Figure 11) clearly shows the MWWTP grouping containing the highest triclosan concentrations and

median. This is similar to results from other studies which emphasized that MWWTP were the most important source of triclosan in the environment (MacLeod and Wong 2010, Kimura et al. 2014).

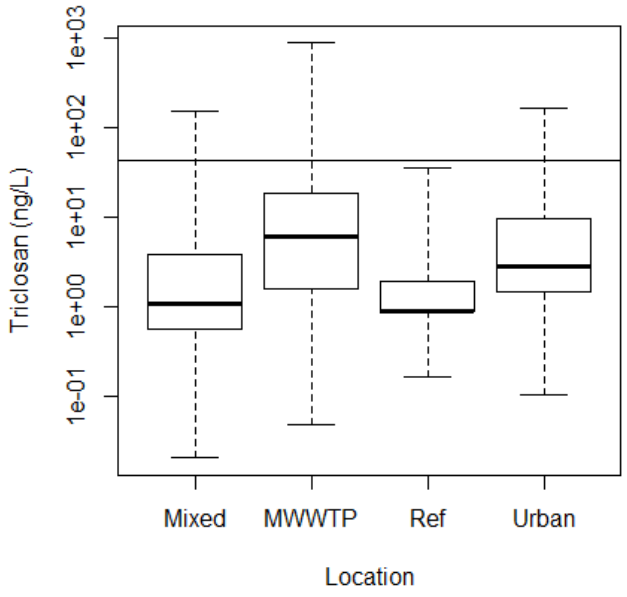

Figure 11. Triclosan concentrations per major river groupings (whereas Mixed (rural and mixed development), MWWTP (Municipal Wastewater Treatment Plant associated sites), Ref (reference sites), Urban (Urban sites)).

4.6 Comparison with Previous Studies

Concentrations of triclosan in select Canadian surface waters have been previously reported to range from below detection (0.10 ng L^{-1}) to 691 ng L^{-1} with means from below detection to 168 ng L^{-1} (HC and EC 2012). The results here (6.1 to 874 ng L^{-1}) fall within that range, though there are some noticeable differences (Table 4). The median triclosan concentration for downstream Wascana creek collected in 2002-03 (168 ng L^{-1}) is higher than the median concentration detected in this study (144 ng L^{-1}, Table 4). Internal Environment Canada data from the Hamilton Harbour sites in 2003-2004 indicate median triclosan concentrations ranging from 10.7 to 230 ng L^{-1} . The median triclosan concentrations have decreased two to four fold depending on the sites in Hamilton Harbour (6.01 to 57.6 ng L^{-1}). Median triclosan concentrations in both Grand River and Thames River downstream seem to have declined

from earlier studies (22.1 and 11 ng L^{-1} respectively), however maximum concentrations for this study were higher than the previous study at the same sites in 2003-2004 (Table 4). These data appear to indicate that triclosan concentrations are declining in Canada. It is important, however, to note that the analytical methods for the unpublished Environment Canada data and most other studies conducted before 2010 used gas chromatography (Table 4) while the AXYS and ECCC (post 2015) methods used liquid chromatography. Therefore, direct comparisons are speculative.

The range of values from this study are within the ranges (nd- 2300 ng L^{-1}) published in past studies from other locations in the world as well (Table 4). It is especially interesting to compare our results to those of the meta-analysis of world-wide values by Brausch and Rand (2011), which had a median value of 48.1 ng L^{-1} at sampling sites downstream of MWWTPs. Wascana Creek (downstream) has a greater median value (144 ng L^{-1}) and Dicks Creek has a median of 62.6 ng L^{-1} but it is based on only three detections, one of which was close to the detection limit. The highest concentrations of triclosan in this study are very similar to those from studies in Japan and China and higher than studies conducted in Australia, Brazil, and Switzerland (Table 4).

Table 4 Comparative average triclosan concentrations

Date	**Details (locations, analytical method)**	**Median Triclosan (ng L^{-1})**	**Range of Triclosan (ng L^{-1})**	**Reference**
Canadian Data				
2002-2003	Wascana downstream	168	12-602	ECCC Pers comm 2011
2012-2016	Wascana downstream	144	13.8-874	This study
2003-2004	Hamilton Harbour 1001 (GC-MS)	14.8	8.07-50.7	Unpublished ECCC data
2012-2016	Hamilton Harbour 1001 (GC-MS)	5.5	5.5-122	This study
2003-2004	Hamilton Harbour 909 (GC-MS)	10.7	5.78-15.6	Unpublished ECCC data
2012-2016	Hamilton Harbour 909 (GC-MS)	6	6-6.1	This study
2003-2004	Hamilton Harbour 914 (GC-MS)	230	13.8-626	Unpublished ECCC data
2012-2016	Hamilton Harbour 914 (GC-MS)	57.6	8.9-268	This study
2003-2004	Hamilton Harbour 926 (GC-MS)	18.75	5.89-49.2	Unpublished ECCC data
2012-2016	Hamilton Harbour 926 (GC-MS)	15.2	7.9-105	This study
2003-2004	Grand River downstream (GC-MS)	26.25	7.17-105	Unpublished ECCC data

Date	**Details (locations, analytical method)**	**Median Triclosan (ng L^{-1})**	**Range of Triclosan (ng L^{-1})**	**Reference**
2012-2016	Grand River downstream (GC-MS)	22.1	5.9-137	This study
2003-2004	Thames River downstream (GC-MS)	40	10-52	Unpublished ECCC data
2012-2016	Thames River downstream (GC-MS)	11	7.5-193	This study
International Data				
1999-2000	United States; 139 steams (GC-MS)	40	50-2300	Koplin et al 2012
1998-2001	Rivers and lakes in Switzerland (GC-MS)	10	1.4-74	Lindström et al 2002
1980-2010	Meta analysis of surface water samples (n=710)	48	<0.1-2300	Brausch and Rand 2011
2004-2005	Australia; 5 rivers (GC-MS)	n/a	14-75	Ying and Kookana 2007
2010-2011	Brazil; 7 rivers (LC-MS)	n/a	2.2-66	Montagner et al 2014
2010-2011	Japan; 13 rivers and streams (GC-MS)	n/a	nd-177	Kimura et al 2014
2011	China; 13 rivers (LC-MS)	67.5	32-294	Wang et al 2014

5.0 Conclusions

Triclosan was detected in 226 out of 918 samples from 44 sites in this large-scale study of high risk sites within the Canadian federal water quality monitoring network. Concentrations ranged from 5.2 to 874 ng L^{-1}, with a mean of 54.23 ng L^{-1} for all the detections. However, using the entire dataset (including censored data with the Kaplan Meier model) resulted in a mean of 17.95 ng L^{-1} and a standard deviation of 52.84 ng L^{-1}. Triclosan in samples from only one site out of 44 exceeded the Federal Environmental Quality Guideline (FEQG) for triclosan established by ECCC (2017). That site (Wascana-downstream) is medium size watercourse which receives effluent from the Regina MWWTP. At that site triclosan measurements were above the FEQG on three different occasions, all of which were during times of low (or no) river flow. Other sites located downstream of MWWTPs also had higher concentrations compared with upstream. The presence and size of treatment plants in relation to the receiving waterbody flow appear to influence both the frequency of detection and triclosan concentrations at downstream sites. Nevertheless, triclosan was detected at a range of reference, rural and urbanized sites as well. Overall, triclosan was detected at 86% of the monitored sites, and was above detection in 25% of samples, indicating that it is commonly found in Canadian surface waters and that sources other than wastewater treatment plants (septic systems, combined sewer/ storm water, cracked sewer pipes) should not be overlooked.

References

AXYS, 2014. Summary of AXYS method MLA-083 Rev 01 Ver 02: Analytical procedure for the determination of triclosan in effluent and other environmental aqueous samples. MSU-083 Rev 04. 23-Jan-2014.

Brausch, J.M., Rand, G.M., 2011. A review of personal care products in the aquatic environment: environmental concentrations and toxicity. Chem. 82, 1518-1532.

Canadian Environmental Protection Act, 1999. *Persistence and Bioaccumulation Regulations*, P.C. 2000-348, 29 March 2000, SOR/2000-107. Canada Gazette, Part II, vol. 134, no 7, p607-612.

Environment and Climate Change Canada (ECCC), Health Canada (HC). 2016. Environment and Climate Change Canada, Health Canada. Assessment report: triclosan. Gatineau (QC).

Environment and Climate Change Canada (ECCC). December 2017. Canadian Environmental Protection Act, 1999. Federal Environmental Quality Guidelines. Triclosan. Ottawa, Canada. https://www.canada.ca/content/dam/eccc/documents/pdf/pded/triclosan/English%20Canadian%20Environmental%20Protection%20Act%201999%20Federal%20Environmental%20Quality%20Guidelines.pdf

EPA, 2007, Method 1694: Pharmaceuticals and personal care products in water, soil, sediment and biosolids by HPLC/MS/MS. EPA-821-R-08-002.

Halden, R.U., 2014. On the need and speed of regulating triclosan and triclocarban in the United States. Env. Sci. Tech. 48, 3603-3611.

Helsel, D.R. 2012. Statistics for censored environmental data using minitab and R. Wiley.

Health Canada (HC) and Environment Canada (EC), 2012. Preliminary Assessment. Triclosan. Chemical Abstracts Service Number 3380-34-5. March 2012. Available at: http://www.ec.gc.ca/ese-ees/default.asp?lang=En&n=6EF68BEC-1.

Kimura, K., Kameda, Y., Yamamoto, H., Nakada, N., Tamura, I., Miyazaki, M., Masunaga, S., 2014. In Press. Occurrence and preservatives and antimicrobials in Japanese rivers. Chemo. 107, 393-9.

Lees and Associates, Karen Hurley and Associates, Hudema Consulting Group Limited, Dayton and Knight Ltd. 2002. Still Creek Rehabilitation and Enhancement Study. City of Vancouver.

Lindström, A., Buerge, I.J., Poiger, T., Bergqvist, P.A., Muller, M.D., Buser, H.R., 2002. Occurrence and environmental behavior of the bactericide triclosan and its methyl derivative in surface waters and in wastewater. Env. Sci. Tech. 36,2322-2329.

Macleod, S.L., Wong, C.S., 2010. Loadings, trends, comparisons, and fate of achiral and chiral pharmaceuticals in wastewaters from urban tertiary and rural aerated lagoon treatments. Water Res. 44, 533-544.

Montagner, C.C., Jardim, W.F., Von der Ohe, P.C., Umbuzeiro, G.A., 2014. Occurrence and potential risk of triclosan in freshwaters of Sao Paulo, Brazil – the need for regulatory actions. Env. Sci. Poll. Res. 21,1850-1858.

NICNAS, 2009. Priority Existing Chemical Assessment Report No 30. Triclosan. January 2009. Australian Government, Department of Health and Ageing.

Québec. 2013. Affaires Municipales, Region et Occupation du territoire. Liste des stations d'épuration. 2013-04-05.

Veldhoen, N., R.C. Skirrow, H. Osachoff, H. Wigmore, D.J. Clapson, M.P. Gunderson, G. van Aggelen and C.C. Helbing. 2006. The bactericidal agent triclosan modulates thyroid hormone–associated gene expression and disrupts postembryonic anuran development. Aquat. Toxicol. 80: 217–227.

Waiser, M.J., Humphries, D., Tumber, V., Holm, J. 2011. Effluent-dominated streams. Part 2: Presence and possible effects of pharmaceuticals and personal care products in Wascana Creek, Saskatchewan, Canada. Env. Toxicol. Chem. 30(2), 508-519.

Wang, X.K., Jiang, X.J., Wang, Y.N., Sun, J., Wang, C., Shen, T.T., 2014. Occurrence, distribution, and multi-phase partitioning of triclocarban and triclosan in an urban river receiving wastewater treatment plants effluent in China. Env. Sci. Poll. Res. 21(11), 7065-7074.

Ying, G.G., Kookana, R.S., 2007. Triclosan in wastewaters and biosolids from Australian wastewater treatment plants. Env. Intl. 33, 199-205.