

# Organochlorine Contaminant, Triclosan Leads to Increased Levels of Trihalomethanes in Drinking Water Sources across the United States

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**How to cite this paper:** Wilburn, W., Guha, S., & Beni, R. (2023). Organochlorine Contaminant, Triclosan Leads to Increased Levels of Trihalomethanes in Drinking Water Sources across the United States. *Voice of the Publisher*, 9, 150-172.

<https://doi.org/10.4236/vp.2023.93014>

**Received:** May 24, 2023

**Accepted:** September 18, 2023

**Published:** September 21, 2023

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## Abstract

Organochlorine contaminants, such as Triclosan (TCS), are present in drinking water sources across the United States. Since TCS was developed in the late 1960s, antimicrobial compounds have been widely used as multipurpose ingredients in everyday consumer products, can be ingested or absorbed through the skin, and are found in human plasma, breast milk, and urine samples. Monitored by the United States Toxic Substances Control Act, TCS production was limited to 1 million pounds per year, yet by 1998 production of TCS steadily increased from 1 million pounds to 10 million, with an estimated production of approximately 14 million globally by 2011. Studies have shown that the expanded use of antimicrobial agents causes them to be found and remain suspended in the ecosystem, most notably the soil and watersheds. Research has shown emerging concerns related to the overuse of TCS, such as dermal irritations, higher incidence of antibacterial-related allergies, microbial resistance, endocrine system disruptions, altered thyroid hormone activity, metabolism, and tumor metastasis and growth, with overexposure playing a role in inflammatory responsiveness, which could cause adverse outcomes and is associated with numerous pathologies, including cardiovascular disease and several types of cancers. To understand the impact of the overuse of TCS-containing products on water quality before the Food and Drug Administration (FDA) began to require pre-market approval, we have analyzed the data reported between March 2005 and 2015 by Consumer Confidence Report (CCR) on the levels of total trihalomethanes (TTHM), such as chloroform, a product of free chlorine added to TCS in the metropolitan areas primary water sources across the United States, as they correlated to increased production of antibacterial agent, TCS. Our study concluded that increased

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use of products containing the antimicrobial agent TCS contributes to higher levels of total organochlorine contaminant, trichloromethane, leading to an increase in TTHM levels recorded annually on water quality reports.

## Keywords

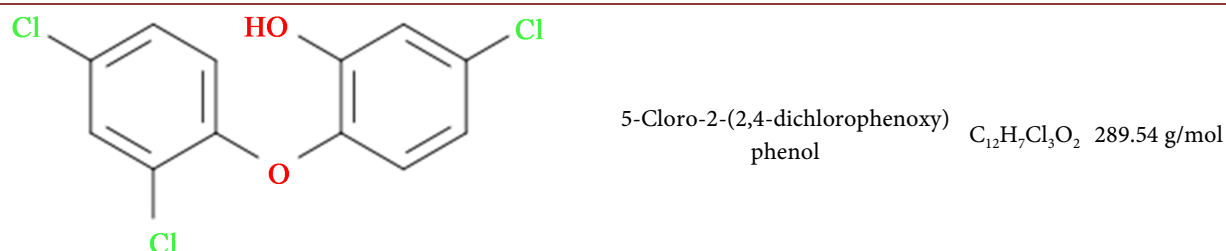
Organochlorine Contaminants, Triclosan, Trihalomethane, Chloroform, Water Quality

## 1. Introduction

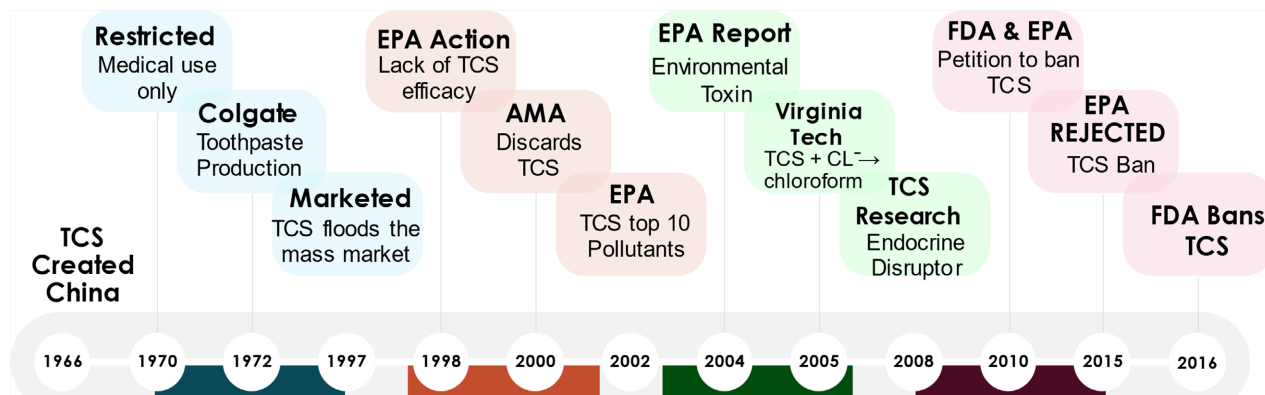
Triclosan (TCS), a chlorinated aromatic compound containing phenol and ether functional groups, is an antimicrobial and antibacterial agent in many consumer products, notably hand sanitizers and aerosol sprays. The molecular structure of TCS is displayed in **Table 1**.

TCS was first created and patented in 1964 by the Swiss to be used as surgical scrubs by hospitals and healthcare workers but entered worldwide production in 1997 as antimicrobial compounds (**Figure 1**) (Ahn et al., 2008). The United States Toxic Substances Control Act monitored the production of TCS, limiting production to approximately 1 million pounds per year, yet by 1998 production of TCS steadily increased from 1 million pounds to 10 million pounds, with an estimated global production of TCS in 2011 of approximately 14 million pounds

**Table 1.** Structure, names, and properties of triclosan.



(Adolfsson-Erici et al., 2002; Bhargava & Leonard, 1996; Perencevich et al., 2001; Witorsch et al., 2014).



**Figure 1.** Historical timeline for triclosan.

(Fang et al., 2010; United States Food and Drug Administration, HHS, 2013, 2016; Perencevich et al., 2001). In 2011 United States consumers spent nearly 1 billion dollars annually on products containing TCS (Statista, 2019).

TCS use has expanded from everyday personal hygiene products such as soaps, mouthwash, toothpaste, beauty aids, cleaning supplies, and pesticides (Boyce & Pittet, 2002; Thompson et al., 2005), becoming a significant component of other marketed consumer goods, such as kitchen utensils, trash bags, toys, diapers, bedding, and socks (Boyce & Pittet, 2002; Thompson et al., 2005). By 2000, TCS had been added to more than 2000 daily used consumer products (Weatherly & Gosse, 2017). Between September 2008 and September 2009, products containing concentrations of 3.5 to 17 mM of TCS as the active antimicrobial ingredient sold at a rate of 278 million 16 oz units totaling \$886 million in total sales resulting in annual consumption of 132 million liters (United States Food and Drug Administration, HHS, 2013; Weatherly & Gosse, 2017). By 2010, TCS was found in 93% of liquid, gel, bar, or foam soaps (United States Food and Drug Administration, HHS, 2013, 2016; Weatherly & Gosse, 2017). Some research suggests that antimicrobial hand soaps containing TCS slightly but significantly reduce bacteria compared to plain soap (Rover & Leu-Wai-See, 2014).

The widespread use of TCS-containing products has given rise to the contamination of aquatic environments, most notably watersheds (Bhargava & Leonard, 1996; Kolpin et al., 2002; Singer et al., 2002). The EPA regulates TCS as a pesticide that is generally acceptable on solid surfaces, while the FDA regulates TCS as a drug when used in personal care items (Halden et al., 2017). As of 2017, the United States Environment Protection Agency (EPA) enacted five registration restrictions for using TCS as an active antimicrobial ingredient added to products used to slow or deter the growth of bacteria, fungi, and mildew. This commercial, institutional, and industrial restriction allows TCS use in fire hoses, dye vats, conveyor belts, and ice-making equipment, along with direct application to commercial Heating, Ventilation, and Air Conditioning (HVAC) coils to prevent microbial growth that contributes to their degradation (Halden et al., 2017; Thomaidi et al., 2017; Weatherly & Gosse, 2017).

### 1.1. Triclosan Toxicology

Organochlorine compounds are not acutely toxic to mammals (Schweizer, 2001; Adolfsson-Erici et al., 2002; Latch et al., 2005; Robertshaw & Leppard, 2007), but *in vitro* studies indicate that TCS may disturb metabolic systems and hormone homeostasis at a low concentration of 0.03 mg/L (0.1  $\mu$ M) or 0.00003% (Hanioka et al., 1997; Jacobs et al., 2005; Schuur et al., 1998; Veldhoen et al., 2006; Wang et al., 2004). According to the FDA monograph for health care products containing TCS, the recommended limits are up to 3000 mg/L (10.36 mM) or 0.3% in oral care products (toothpaste) and leave-on, dermal care products (deodorant and lotion), 1000 - 4500 mg/L (3.45 - 15.54 mM) or 0.1% - 0.45%, in wash away dermal care products (liquid hand and body soaps), and 10,000 - 30,000 mg/L

(34.5 - 103.6 mM) or 1% - 3% in products used in health care settings (Cosmetics Directive, 2014; United States Food and Drug Administration, HHS, 2013, 2016; Lindström et al., 2002).

The levels of TCS that may be present in tissues just beneath the skin following exposure to antimicrobial products that contain as much as 0.3% organochlorine compound could be greater than 10 µM, based on findings of the absorption in human skin samples (Moss et al., 2000). Studies have shown that TCS disrupts the thyroid hormone-dependent characteristics of metamorphosis in frogs (Veldhoen et al., 2006) and has been shown to interfere with thyroid hormone regulation in rats (Crofton et al., 2007). Researchers found that tadpoles treated with low levels of organochlorine compounds have altered hormone-mediated development in the frog studies, while TCS exposure also disrupted thyroid hormone-associated gene expression (Wang & Tian, 2015; Wang et al., 2020). Studies on frogs and rats have demonstrated that TCS can profoundly affect thyroid hormones (Wang et al., 2020). Even though several recent studies have found concerns that TCS is an endocrine disruptor, the complete relevance of the extent to humans is still unknown (Bedoux et al., 2012). Recently, researchers at UC Davis found that TCS elevates calcium levels in cells, potentially affecting neurological function and neurodevelopment; while also impairing mitochondrial function in mammalian cells (Benotti et al., 2009; Ma et al., 2018).

## 1.2. Triclosan Exposure & Inflammation

Since the discovery that inflammation can play a role in tumor development, scientists have shown that inflammation is present in the same areas as tumor cells, showing a connection between pro-inflammatory cells and cancer cells (Balkwill & Mantovani, 2001). Inflammation can become chronic under the wrong circumstances, leading to inflammatory and autoimmune diseases. Acute inflammation occurs when tissue repair is required due to injury and helps to prevent infections (Balkwill & Mantovani, 2001). Chronic inflammation occurs when pro-inflammatory and various mediators stimulate immune cells from lymphoid and myeloid lineages in blood vessels (Hermouet et al., 2015). Inflammation can appear before and during malignant tumor growth and metastasis due to immune cell involvement in inflammation (Hermouet et al., 2015). When immune responsiveness levels are inappropriately elevated, it causes chronic inflammation that can increase tumor invasiveness (Voronov et al., 2003). Any organochlorine, TCS-induced increase in pro-inflammatory responsiveness could result in chronic inflammation, which is associated with numerous pathologies, including cardiovascular disease and several types of cancers (Amin et al., 2020; Dinarello, 2009; Kaneko et al., 2019; Lewis et al., 2006; Shirazi et al., 2017; Singh et al., 2019; Voronov et al., 2003). Additionally, TCS was detected in 75% of 2517 human urine samples at concentrations of 2.4 - 3790 µg/L (Calafat et al., 2008), in 61% of 90 urine samples from age 6 - 8-year-old girls (Wolff et

al., 2007) and in human blood, TCS was detected in the range between 4.1 - 19 ng/g in blood serum samples (Allmyr et al., 2006, 2008). TCS concentrations between 100 - 2100 µg/kg of lipid were detected in 96.8% of 62 breast milk samples (Dayan, 2007), and concentrations of TCS in breast milk were detected between 0.018 to 0.95 ng/g (Allmyr et al., 2006). Lastly, TCS has been found in indoor dust (~1.1 µg/g) (Canosa et al., 2007) and foods (0.02 - 0.15 ng/g) such as dairy products, vegetables, meat, fish, and egg (Allmyr et al., 2006).

## 2. Water Contaminants and Regulations

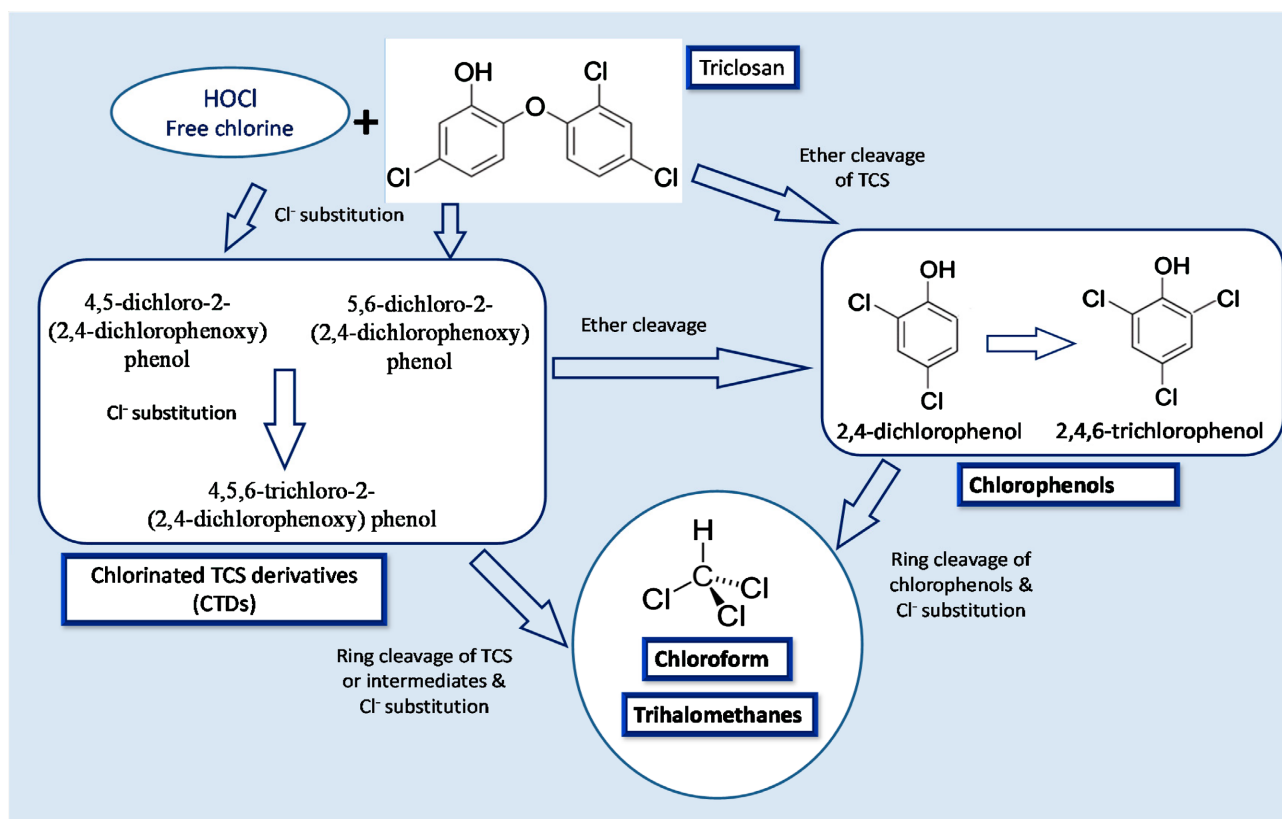
Drinking water quality varies depending on the treatment procedure or condition of the source water, but it must meet US Environmental Protection Agency (EPA) standards and regulations. The underlying importance of water is understood, albeit violations do occur even with EPA standards in place. Once a violation occurs, it must be reported to the EPA, while the EPA is responsible for the CCR for informing the consumer of the water quality (United States Environmental Protection Agency, 2022a). The levels of contaminants found in drinking water have been studied by scientists at various agencies, such as the non-profit corporation Environmental Work Group (EWG). The EWG specializes in determining the links between tested chemical compounds found in a water source and the environmental consequences, even if the levels of the compound are within legal limits assigned by the EPA (Jones et al., 2023). Previous studies in our lab found a correlation between the levels of trihalomethanes (THMs), annual household income, and poverty levels (Guha et al., 2019). Other studies showed water quality disparities regarding the level of heavy metals exposure in the drinking water of Tennessee (Beni et al., 2019). Our primary exposure to environmental hazards is unsafe drinking water.

The EPA is responsible for writing regulations to enforce water quality legislation, such as the National Primary Drinking Water Regulations, the National Primary Drinking Water Regulations Implementation, and the National Secondary Drinking Water Regulations (Baum et al., 2015). However, the EPA only requires the regulations of public drinking water systems that service at least 15 service connections or more than 25 persons (Levin et al., 2002). The Safe Drinking Water Act of 1974 requires public drinking water systems to monitor the presence of certain contaminants at specific intervals of time and at mandatory locations to ensure compliance allowing violations to be reported to the Safe Drinking Water Information System Federal Reporting Services (SDWIS/FED), created in 1995 (Weinmeyer et al., 2017). However, a 2002 EPA audit found that only 62% of violations are ever reported, and states are only required to report a violation, not the contamination levels for water supply systems servicing 100,000 or greater. These protocol violations leave citizens with only the knowledge of a possible violation, not the specifics of the violation (United States Environmental Protection Agency, 2016).

## Contaminants in Drinking Water from Triclosan Use

Many studies have reported the occurrence of TCS and its intermediates in wastewater effluent, watersheds, and soil (Subedi et al., 2014; Karthikraj & Kannan, 2017). According to the EPA, TCS is in the top 10 contaminants of emerging concern for watersheds in the United States (Subedi et al., 2014; Vimalkumar et al., 2019; Abbott et al., 2020). Once organochlorine contaminants enter wastewater treatment plants, free chlorine exposure causes a partial or complete transformation during wastewater treatment processing before being discharged into the environment via effluent and biosolids for land application (Figure 2).

The land application of biosolids, the final products of wastewater treatment plants, can potentially route chloroform into the environment. Organochlorine aerobic biodegradation is relatively slow, with a half-life greater than 165 days (Thelusmond et al., 2018). Although organochlorine contaminants in the effluent and biosolids of water resources recovery facilities are currently not regulated, the public interest has led Metropolitan Water Resources (MWR) and the EPA to monitor chloroform in the influent, effluent and biosolids (Armstrong et al., 2017). A critical comprehensive review of TCS as an environmental contaminant has been recently reported, discussing the causal effect of free chlorine chemical response to organochlorine contaminants increasing bio-solids concentration



(Dhillon et al., 2015).

**Figure 2.** Mechanisms of TCS conversion to its intermediate products: chlorinated TCS derivatives, chloroform, and trihalomethanes.

before land application (Armstrong et al., 2017).

Exposure to THMs can have serious adverse health effects after prolonged exposure to higher levels of contaminants. **Table 2** shows common water contaminants found in the drinking water supply and their relative limits, sources, and potential health risks associated with their exposure. The limits and sources for each contaminant are listed in every CCR, and limit violations are reported by SDWIS/FED annually. Our report will provide timely yet informative results for initial screening on drinking water quality and risk identification that can be incorporated into a water resource management strategy.

### 3. Materials and Methods

All secondary data related to levels of THM concentrations were obtained from the annual water safety report MWR for the metropolitan water plants across the United States. See Districts (**Figure 3; Table 3**) for each metropolitan city for each state in the United States (United States Environmental Protection Agency, 2022b).

#### 3.1. Materials

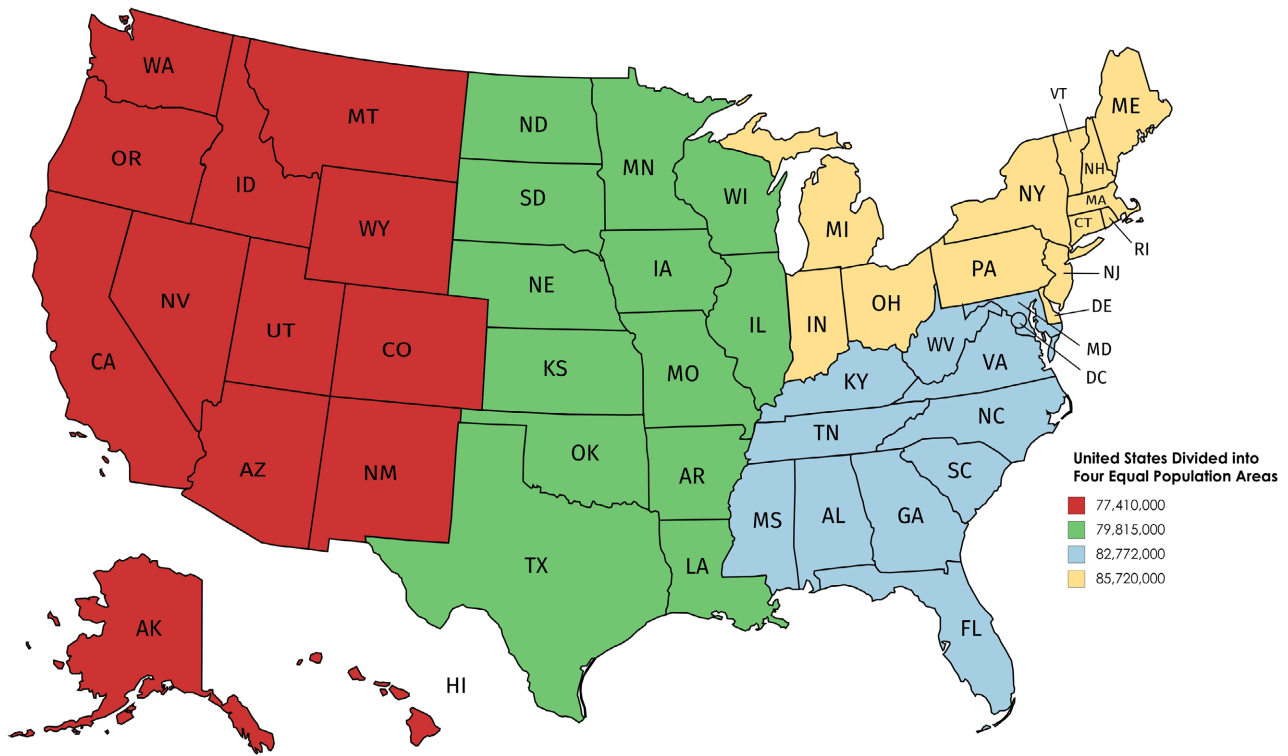
The water quality data was then prepared for descriptive statistical analysis from secondary data related to drinking water quality obtained from the annual water safety reports for the major cities of each state in the United States divided into districts (United States Environmental Protection Agency, 2022a). Data, including median annual household income, was obtained from the United States Census Bureau (United States Census Bureau, 2020). Additional information was collected by contacting water service offices to obtain information not readily available in the annual water safety report.

Tables were generated to record income per capita for each state (provided by the Census Bureau) along with their drinking water sources (provided by the state and local water services departments) and correlated to the levels of contaminants.

**Table 2.** Common drinking water contaminants with their relative limits, sources, and potential health effects.

Contaminants	Limits	Sources	Potential Health Effect
Residual Chlorine	4 mg/L	Disinfectant added to control pathogens	Nervous System Damage, Anemia, Methemoglobinemia
Trihalomethane: chloroform bromodichloromethane dibromochloromethane bromoform	80 mg/L	By-product of water chlorination	Cancer, Liver Disease, Kidney Failure, Nervous System Damage, Bladder Cancer, Colorectal Cancer, Adverse development, and reproductive effects during pregnancy

(United States Environmental Protection Agency, 2016).



(United States Census Bureau, 2020).

**Figure 3.** Map showing the census bureau regions and divisions with state FIPS codes of the United States.

**Table 3.** Census bureau regions and divisions with state FIPS codes regions division for each state in the United States.

WEST	MIDWEST	SOUTH	NORTHEAST
AK—Alaska	IL—Illinois	AL—Alabama	CT—Connecticut
AZ—Arizona	IN—Indiana	AR—Arkansas	DE—Delaware
CA—California	IO—Iowa	FL—Florida	ME—Maine
CO—Colorado	KS—Kansas	GA—Georgia	MD—Maryland
HA—Hawaii	MI—Michigan	KY—Kentucky	MA—Massachusetts
ID—Idaho	MN—Minnesota	LA—Louisiana	NH—New Hampshire
MT—Montana	MO—Missouri	MS—Mississippi	NJ—New Jersey
NV—Nevada	NE—Nebraska	NC—North Carolina	NY—New York
NM—New Mexico	ND—North Dakota	OK—Oklahoma	PA—Pennsylvania
OR—Oregon	OH—Ohio	SC—South Carolina	RI—Rhode Island
UT—Utah	SD—South Dakota	TN—Tennessee	
WA—Washington	WI—Wisconsin	TX—Texas	VT—Vermont
WY—Wyoming		VA—Virginia	
		WV—West Virginia	

(United States Census Bureau, 2020).

The disparities among the average household income in different states and their water quality are shown using multi-variable charts. **Table 3** features the states



divided into districts (West, Midwest, South, and Northeast) as reported by the US Census and the EPA being analyzed in this study. Additional information for each water system was obtained from the EPA website using the Safe Drinking Water Information System (SDWIS) (United States Environmental Protection Agency, 2022c). The SDWIS provided the primary water source, the number of violations, and the population served for each water system.

The median household income, population data, and % of persons in poverty were retrieved from the United States Census Bureau for 2010 and 2020 (United States Census Bureau, 2020) censuses. The data was obtained by accessing the QuickFacts website for the Census Bureau. Since the release of the 2015 census, Quick-Facts has shown the information from the current censuses. The population information is reported for 2010 and 2020.

### 3.2. Statistical Analysis

Statistical data analysis was performed using ANOVA and Student's t-test. ANOVA initially compared data within a given experimental setup (Qualtrics, 2023). A significant ANOVA was followed by a pairwise analysis of control versus exposed data using Student's t-test; a *p*-value of less than 0.05 was considered significant.

## 4. Results and Discussion

According to the 2010 data from the US Census Bureau, the nation's poverty rate rose to 15.1 percent, the highest level in 17 previous years, with the median income dropping by more than two percent from about \$49,445. According to the 2010 US Census, the US had a population of 308.7 million which was a 9.7% increase since the last census in 2000, and an annual income level of \$50,046 before taxes, which was an annual growth rate of 3.1 percent (US Census Report), with approximately 15.1 percent under the federal poverty level (FPL) of the US: The number of people living in poverty increased from 42.9 million to 46.2 million during the same period (United States Census Bureau, 2022).

Based on **Table 4** and **Figure 4**, the West District of the US has an average population of 5.5 million with an average annual salary of \$51,988, 0.12 percent higher than the US annual income. The mean average levels of TTHM [ppb] (M) were  $25.3 \pm 2.0$  ppb with (SD) 56.4 ppb for 2005 and (M)  $62.5 \pm 3.5$  ppb with (SD) 12.2 ppb for 2015, as shown in **Table 5**. A student's T Test showed statistical significance in the increase in the levels of TTHMs between 2005 and 2015 with a *p* value of  $7.72E-08$  (1-tailed) and  $1.54E-07$  (2-tailed). When comparing the averages (n=124) for 2005-2015, the increase in TTHM levels was found to be statistically significant *f*critical 1.8 and *p* value  $1.9E-4$  (2-tailed ANOVA), with an average % increase in the TTHM levels of 156.1 percent from 2005-2015.

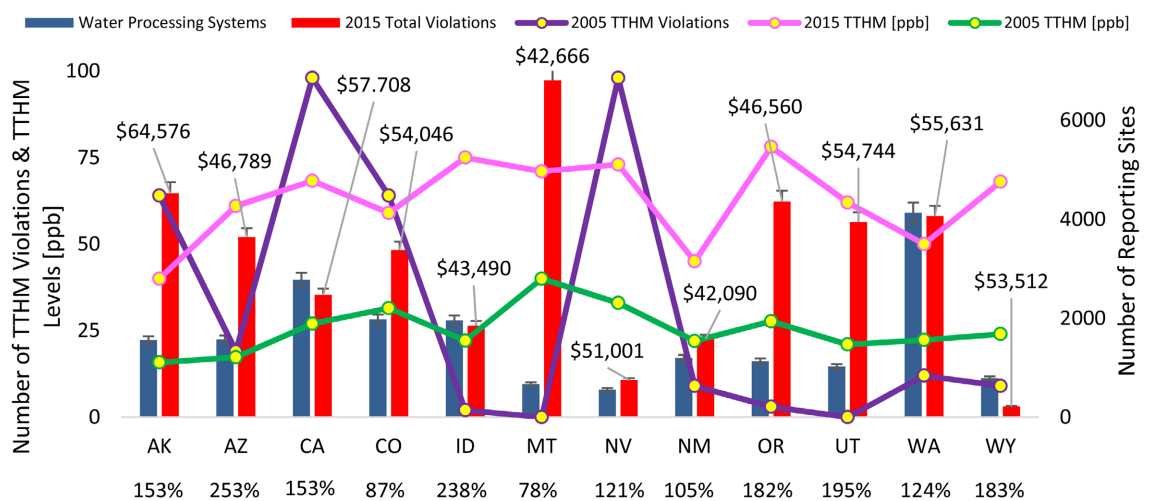
Based on **Table 6** and **Figure 5**, the Midwest District of the US has an average population of 5.6 million with an average annual salary of \$48,004, 4.3 percent lower than the US annual income. The mean average levels of TTHM [ppb] (M)

were  $30.5 \pm 5.3$  ppb with (SD) 18.2 ppb for 2005 and (M)  $58.9 \pm 6.7$  ppb with (SD) 14.7 ppb for 2015, as shown in **Table 7**. A student's T Test showed statistical significance in the increase in the levels of TTHMs between 2005 and 2015

**Table 4.** West District of the United States—US Census Data for the average Population and Annual Income. The US EPA—SDWIS/FED Annual Data for the number of Total Violations, TTHM violations, and TTHM Levels reported by each state of the West District of the United States.

State		2010			2005			2015			
		Census Data			Violations		TTHM	Violations		TTHM	
		Population	Income	Systems	Total	TTHM	[ppb]	Total	TTHM	[ppb]	% Change
AK	Alaska	710,231	\$64,576	1556	3687	64	15.8	4527	42	40.0	153.2%
AZ	Arizona	6,392,017	\$46,789	1573	5018	19	17.3	3637	3	61.0	252.6%
CA	California	37,253,956	\$57,708	7781	2205	98	27.0	2473	103	68.3	153.0%
CO	Colorado	5,029,196	\$54,046	1975	1325	64	31.5	3379	39	59.0	87.3%
HA	Hawaii	1,360,301	\$63,030	130	47	NR	NR	5	NR	NR	NR
ID	Idaho	1,567,582	\$43,490	1957	887	2	22.1	1851	2	75.0	239.4%
MT	Montana	989,415	\$42,666	670	3914	0	40.0	6810	44	71.0	77.5%
NV	Nevada	2,700,551	\$51,001	558	815	198	33.0	752	2	73.0	121.2%
NM	New Mexico	2,059,179	\$42,090	1195	860	9	22.0	1587	13	45.0	104.5%
OR	Oregon	3,831,074	\$46,560	1130	3483	3	27.7	4360	6	78.1	181.9%
UT	Utah	2,763,885	\$54,744	1022	1936	0	21.0	3945	2	62.0	195.2%
WA	Washington	6,724,540	\$55,631	4133	10,399	12	22.3	4069	76	50.0	124.2%
WY	Wyoming	563,626	\$53,512	787	233	9	24.0	215	2	68.0	183.3%

Note. NR means no data recorded (United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).



**Figure 4.** West District of the United States—US Census Data for the average Population and Annual Income. The US EPA—SDWIS/FED Annual Data for the number of Total Violations, TTHM violations, and TTHM Levels reported by each state of the West District of the United States.

**Table 5.** West district of the United States—statistical descriptive data for the averages reported by the states of the west district.

Statistics	2010			2005			2015			
	Census Data			Violations		TTHM	Violations		TTHM	
	Population	Income	Systems	Total	TTHM	[ppb]	Total	TTHM	[ppb]	% Change
Mean (M)	5,534,273	\$51,988	1882.1	2677.6	39.8	25.3	2893.1	27.8	62.5	+156.1%
Standard Error	2.7E+6	2049.2	562.4	775.3	17.1	2.0	546.1	9.7	3.5	16.3
Variance	9.5E+13	5.5E+7	4.1E+6	7.8E+6	3513	47.9	3.9E+6	1139	148.1	3182.6
Standard Deviation (SD)	9.8E+6	7388.5	2027.9	2795.4	56.4	6.9	1968.7	33.8	12.2	56.4

Note. % is the net increase in TTHM levels from 2005-2010 (United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).

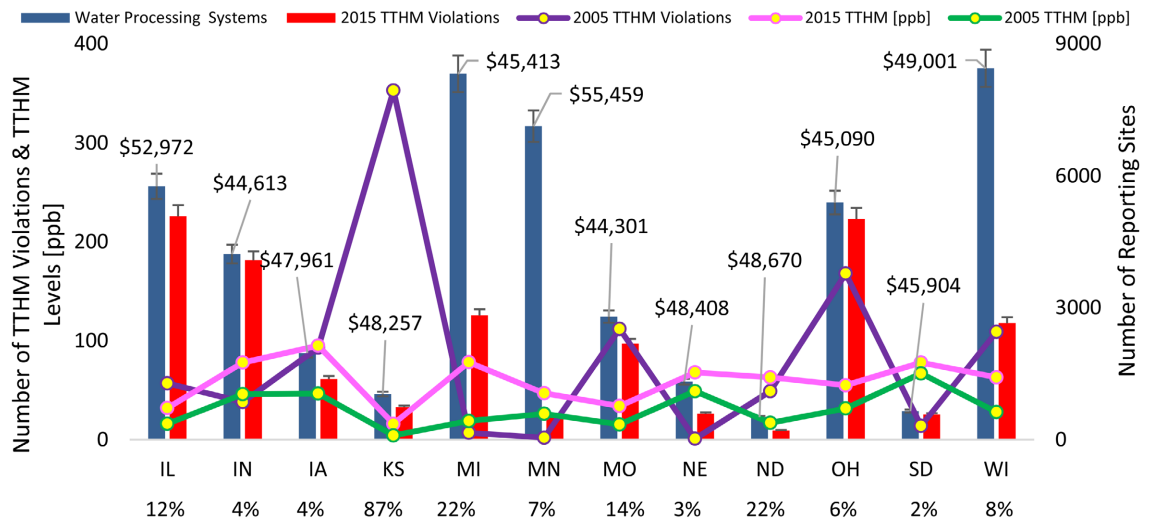
**Table 6.** Midwest district of the United States—US census data for the average population and annual income. The US EPA—SDWIS/FED annual data for the number of total violations, TTHM Violation, and TTHM Levels reported by each state of the midwest district of the United States.

State		2010			2005			2015			
		Census Data			Violations		TTHM	Violations		TTHM	
		Population	Income	Systems	Total	TTHM	[ppb]	Total	TTHM	[ppb]	% Change
IL	Indiana	12,830,632	\$52,972	5761	5077	57	16.1	4305	20	32.0	12.3%
IN	Indiana	6,483,802	\$44,613	4221	4079	38	46.0	2039	5	78.0	3.7%
IA	Iowa	3,046,355	\$52,972	1966	1377	93	46.6	911	5	95.0	4.4%
KS	Kansas	2,853,118	\$44,613	1035	738	353	4.3	382	36	16.0	86.5%
MI	Michigan	9,883,927	\$47,961	11320	2824	7	18.9	2478	1	78.3	21.9%
MN	Minnesota	5,303,925	\$48,257	7128	484	2	26.2	450	37	46.9	6.8%
MO	Missouri	5,988,927	\$45,413	2797	2182	112	15.4	1801	58	34.0	14.3%
NE	Nebraska	1,826,341	\$55,459	1319	588	1	49.0	516	6	68.0	2.8%
ND	N. Dakota	672,591	\$44,301	512	209	49	17.0	375	19	63.0	21.8%
OH	Ohio	11,536,504	\$48,408	5392	5020	168	31.6	2340	39	55.0	5.5%
SD	S. Dakota	814,180	\$48,670	652	571	14	67.0	466	3	78.0	1.7%
WI	Wisconsin	5,686,698	\$45,090	5761	5077	57	16.1	4305	20	32.0	8.0%

(United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).

with a *p* value of 3.15E−05 (1-tailed) and 6.03E−05 (2-tailed). When comparing the averages (n=119) for 2005-2015, the increase in TTHM levels was found to be statistically significant *f* critical 1.97 and *p* value 3.40E−21 (2-tailed ANOVA), with an average % increase in the TTHM levels of 15.8 percent from 2005-2015.

Based on **Table 8** and **Figure 6**, the South District of the US has an average population of 7.7 million with an average annual salary of \$43,244, 15.7 percent lower than the US annual income. The mean average levels of TTHM [ppb] (M) were 36.1 ± 2.6 ppb with (SD) 9.6 ppb for 2005 and (M) 67.6 ± 2.9 ppb with (SD)



**Figure 5.** Midwest District of the United States—US Census Data for the average Population and Annual Income. The US EPA—SDWIS/FED Annual Data for the number of Total Violations, TTHM Violation, and TTHM Levels reported by each state of the Midwest District of the United States.

**Table 7.** Midwest District of the United States—Statistical Descriptive Data for the Averages Reported by the States of the Midwest District.

Statistics	2010			2005			2015			
	Census Data			Violations	TTHM	Violations	TTHM			% Change
	Population	Income	Systems	Total	TTHM [ppb]	Total	TTHM [ppb]	TTHM [ppb]		
Mean (M)	5,577,250	\$48,004	4462.3	2150.0	83.6	30.5	1495.8	24.7	58.9	+15.8%
Standard Error	1.2E+6	979.9	1122.2	517.1	28.7	5.3	348.8	6.5	6.7	6.7
Variance	1.7E+13	1.2E+7	1.5E+7	3.2E+6	9947	330.8	1.5E+6	508.6	533.5	543.7
Standard Deviation (SD)	4.1E+6	3,394.5	2470.0	1138.2	63.4	11.6	767.8	14.3	14.7	2.3

Note. % is the net increase in TTHM levels from 2005-2010 (United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).

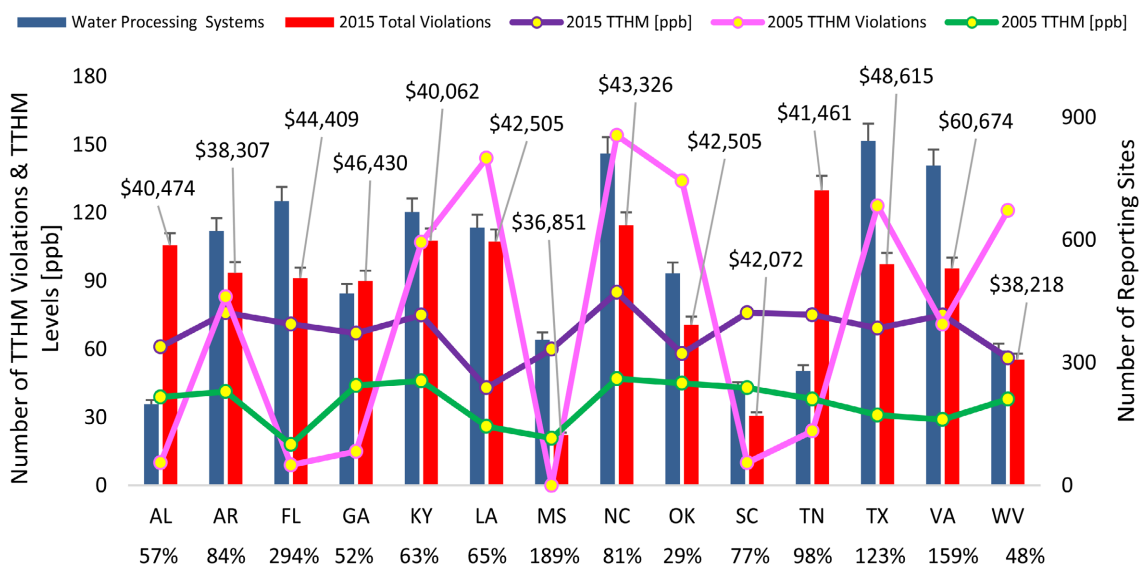
**Table 8.** South District of the United States—US Census Data for the average Population and Annual Income. The US EPA—SDWIS/FED Annual Data for the number of Total Violations, TTHM Violation, and TTHM Levels reported by each state of the South District of the United States.

State		2010			2005			2015			
		Census Data			Violations	TTHM	Violations	TTHM			% Change
		Population	Income	Systems	Total	TTHM [ppb]	Total	TTHM [ppb]	TTHM [ppb]		
AL	Alabama	4,779,736	\$40,474	199	377	10	38.9	587	40	61.0	56.8%
AR	Arkansas	2,915,918	\$38,307	622	804	83	41.3	620	42	76.0	84.0%
FL	Florida	18,801,310	\$44,409	1695	1976	9	18.0	2407	141	71.0	294.4%
GA	Georgia	9,687,653	\$46,430	1469	1995	15	44.0	2600	15	67.0	52.3%
KY	Kentucky	4,339,367	\$40,062	668	643	107	46.0	898	22	75.0	63.0%
LA	Louisiana	4,533,372	\$42,505	630	779	344	26.0	1596	169	43.0	65.4%

Continued

MS	Mississippi	2,967,297	\$36,851	356	863	0	20.8	123	4	60.0	189.2%
NC	N. Carolina	9,535,483	\$43,326	5211	10,437	354	47.0	3636	103	85.0	80.9%
OK	Oklahoma	3,751,351	\$42,072	2519	1905	634	45.0	2393	367	58.0	28.9%
SC	S. Carolina	4,625,364	\$42,018	240	365	10	43.0	170	8	76.0	76.7%
TN	Tennessee	6,346,105	\$41,461	280	741	24	38.0	721	3	75.1	97.6%
TX	Texas	25,145,561	\$48,615	5442	4300	923	31.0	10,541	320	69.2	123.2%
VA	Virginia	8,001,024	\$60,674	2282	3053	71	29.0	2530	11	75.0	158.6%
WV	W. Virginia	1,852,994	\$38,218	330	3852	121	38.0	307	10	56.1	47.6%

(United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).



**Figure 6.** South District of the United States—US Census Data for the average Population and Annual Income. The US EPA—SDWIS/FED Annual Data for the number of Total Violations, TTHM Violations, and TTHM Levels reported by each state of the South District of the United States.

**Table 9.** South District of the United States—Statistical Descriptive Data for the Averages Reported by the States of the South District.

Statistics	2010			2005			2015			
	Census Data			Violations	TTHM	Violations	TTHM			
	Population	Income	Systems	Total	TTHM [ppb]	Total	TTHM [ppb]	% Change		
Mean (M)	7,663,038	\$43,244	1567.4	2292.1	193.2	36.1	2080.6	89.6	67.7	+101.3%
Standard Error	1.8E+6	1588.0	472.3	713.9	74.6	2.6	715.4	32.1	2.9	19.0
Variance	4.4E+13	4.0E+7	3.1E+6	7.1E+6	7.8E+4	91.5	7.2E+6	1.4E+4	119.6	5029.4
Standard Deviation (SD)	6.6E+6	5,942.4	1767.3	2671.4	279.3	9.6	2676.8	120.3	10.9	70.9

Note. % is the net increase in TTHM levels from 2005-2010 (United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).

**Table 10.** Northeast District of the United States—US Census Data for the average Population and Annual Income. The US EPA—SDWIS/FED Annual Data for the number of Total Violations, TTHM violations, and TTHM Levels reported by each state of the Northeast District of the United States.

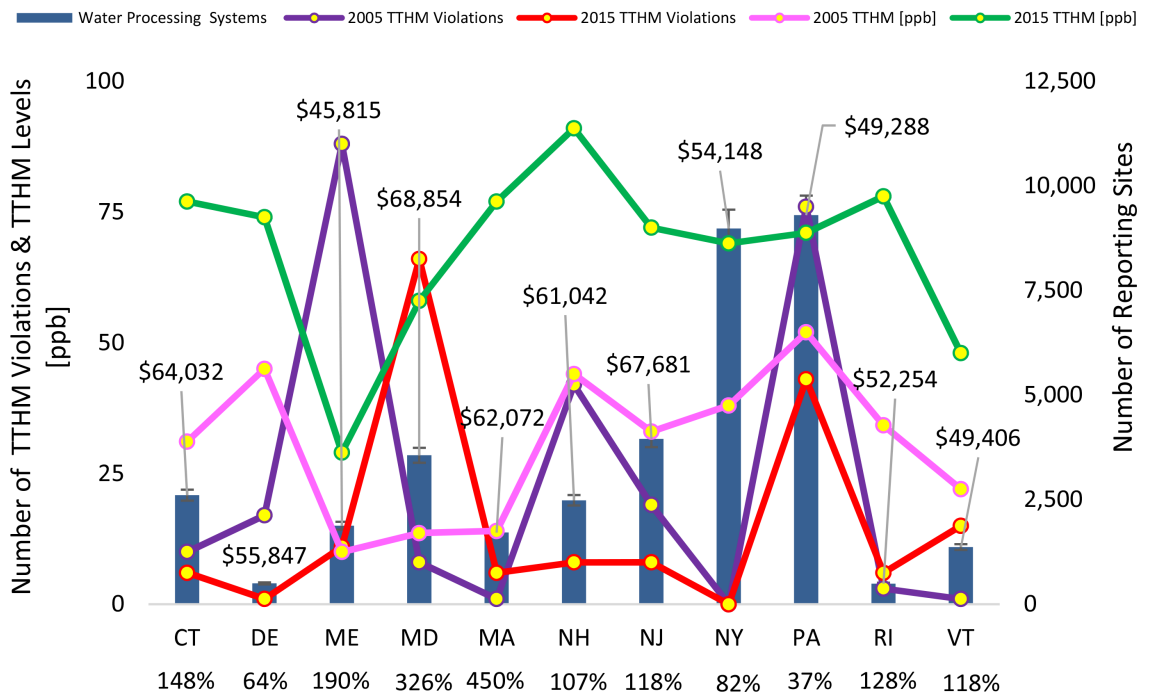
State		2010			2005			2015			% Change
		Census Data			Violations		TTHM	Violations		TTHM	
		Population	Income	Systems	Total	TTHM	[ppb]	Total	TTHM	[ppb]	
CT	Connecticut	3,574,097	\$64,032	2605	5879	10	31.1	3445	6	77	147.6%
DE	Delaware	897,934	\$55,847	496	121	17	45	125	1	74	64.4%
ME	Maine	1,328,361	\$45,815	1875	2411	88	10	1105	11	29	190.0%
MD	Maryland	5,773,552	\$68,854	3559	736	8	13.6	1239	66	58	326.5%
MA	Massachusetts	6,547,629	\$62,072	1712	180	1	14	2314	6	77	450.0%
NH	New Hampshire	1,316,470	\$61,042	2483	1100	42	44	926	8	91	106.8%
NJ	New Jersey	8,791,894	\$67,681	3950	2296	19	33	3018	8	72	118.2%
NY	New York	19,378,102	\$54,148	8979	7189	474	38	5886	NR	69	81.6%
PA	Pennsylvania	12,702,379	\$49,288	9298	13,734	76	52	19,031	43	71	36.5%
RI	Rhode Island	1,052,567	\$52,254	490	88	3	34.2	164	6	78	128.1%
VT	Vermont	625,741	\$49,406	1367	642	1	22	880	15	48	118.2%

(United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).

10.9 ppb for 2015, as shown in **Table 9**. A student's T Test showed statistical significance in the increase in the levels of TTHMs between 2005 and 2015 with a  $p$  value of  $7.72E-08$  (1-tailed) and  $1.54E-07$  (2-tailed). When comparing the averages ( $n=119$ ) for 2005-2015, the increase in TTHM levels was found to be statistically significant  $f$  critical 1.95 and  $p$  value  $1.1E-19$  (2-tailed ANOVA), with an average % increase in the TTHM levels of 101.3 percent from 2005-2015.

Based on **Table 10** and **Figure 7**, the Northeast District of the US has an average population of 5.6 million with an average annual salary of \$57,313, 14.5 percent higher than the US annual income. The mean average levels of TTHM [ppb] (M) were  $30.6 \pm 4.2$  ppb with (SD) 14.1 ppb for 2005 and (M)  $67.7 \pm 5.1$  ppb with (SD) 16.9 ppb for 2015, as shown in **Table 11**. A student's T Test showed statistical significance in the increase in the levels of TTHMs between 2005 and 2015 with a  $p$  value of  $1.92E-06$  (1-tailed) and  $3.84E-06$  (2-tailed). When comparing the averages ( $n=108$ ) for 2005-2015, the increase in TTHM levels was found to be statistically significant  $f$  critical 1.99 and  $p$  value  $4.3E-9$  (2-tailed ANOVA), with an average % increase in the TTHM levels of 160.7 percent from 2005-2015.

After compiling the secondary data, population size, annual income, and water quality for all 50 states in four districts of the US (**Tables 4-11** and **Figures 4-7**). The average mean (M) household income of  $\$49,755 \pm 1140$ , standard deviation (SD) of \$8060, which was statistically significant  $df$  of 99, and  $p$  value of  $2.8E-64$  (2-tailed ANOVA). The average mean number of TTHM violations



**Figure 7.** Northeast District of the United States—US Census Data for the average Population and Annual Income. The US EPA—SDWIS/FED Annual Data for the number of Total Violations, TTHM violations, and TTHM Levels reported by each state of the Northeast District of the United States.

**Table 11.** Northeast District of the United States—Statistical Descriptive Data for the Averages Reported for the States of the Northeast District.

Statistics	2010			2005			2015			% Change
	Census Data			Violations	TTHM	Violations	TTHM	TTHM		
	Population	Income	Systems	Total	TTHM [ppb]	Total	TTHM [ppb]	TTHM [ppb]		
Mean (M)	5,635,339	\$57,313	3346.7	3125.1	67.2	30.6	3466.6	17.0	67.6	+160.7%
Standard Error	1.8E+6	2372.7	923.9	1281.8	41.7	4.2	1638.7	6.6	5.1	37.0
Variance	3.6E+13	6.2E+7	9.4E+6	1.8E+7	1.9E+4	198.1	2.9E+7	433.1	287.3	1.5E+4
Standard Deviation (SD)	6.0E+6	7869.4	3064.2	4251.4	138.3	14.1	5435.0	20.8	16.9	122.8

Note. % is the net increase in TTHM levels from 2005-2010 (United States Census Bureau, 2022; United States Environmental Protection Agency, Archived SDWIS/FED, CCR, 2022c, 2022d).

**Table 12.** United States—Statistical Descriptive Data for the Averages Reported for the West, Midwest, South, and Northeast Districts.

Statistics	2010			2005			2015			% Change
	Census Data			Violations	TTHM	Violations	TTHM	TTHM		
	Population	Income	Systems	Total	TTHM [ppb]	Total	TTHM [ppb]	TTHM [ppb]		
Mean (M)	6,162,876	\$49,755	2735.4	2541.5	100.5	30.9	2456.4	42.8	64.3	+107.1%
Standard Error	9.7E+5	1139.9	413.3	408.7	25.5	1.9	441.6	10.6	2.3	13.3
Variance	4.7E+13	6.5E+7	8.5E+6	8.3E+6	3.2E+4	168.7	9.8E+6	5405.6	262.3	8708.0
Standard Deviation (SD)	6.8E+6	8060.4	2922.6	2890.1	178.7	12.9	3122.9	73.5	16.2	93.3

and levels of TTHM [ppb] (M)  $100.5 \pm 25.5$  with (SD) 178.7, and (M)  $30.9 \pm 1.9$  ppb (SD) 12.9 ppb for 2005; (M)  $42.8 \pm 10.6$  with (SD) 73.5, and (M)  $64.3 \pm 2.3$  ppb (SD) 16.2 ppb for 2015, as shown on **Table 12**. A student's T Test showed statistical significance in the number of TTHM violations reported and the increase in the levels of TTHMs between 2005 and 2015 with a  $p$  value of 0.002 and  $3.05E-23$  (1-tailed) and 0.005 and  $6.10E-23$ , (2-tailed), respectively. When comparing the averages (n=788) for 2000-2015, the increase in TTHM levels was found to be statistically significant  $f$  critical 3.1 and  $p$  value  $9.6E-39$ , with an average % increase in the levels of TTHM was 107.1 percent from 2005-2015 (2-tailed ANOVA).

## 5. Conclusion

Our review assesses the negative impacts on water quality caused by the 312% increase in the use of antiviral products containing the organochlorine TCS (Statista, 2019). Our use of antimicrobial hand sanitizer imported from China spiked in the late 1990s, and on average, consumers used antimicrobial products 9 times per day with an added 25% increase in TCS. Since the mid-1990s, the addition of the organochlorine chemical, Triclosan, which has been incorporated into many consumer products, has significantly increased. Most antibacterial compounds are added to consumer products without a fully encompassed toxicological profile. Since its original use in hospital settings in 1972, Triclosan is an antimicrobial agent incorporated into various consumer products, including soaps, hand sanitizers, toothpaste, and mouthwash. In 1977, TCS production (covered by the United States Toxic Substances Control Act) was between 0.5 and 1 million pounds per year, with production increasing to 10 million pounds in 1998 (United States Food and Drug Agency, HHS, 2013). Since it was estimated that the production of TCS was 14 million pounds by 2011, with an annual consumption of 132 million liters (United States Food and Drug Agency, HHS, 2013), a comprehensive study is essential to assess the contamination of aquatic environments, most notably drinking water.

Our research showed that all states had varying increases in levels of TTHM in major metropolitan water plants and water sources. All states had statistically significant variability for averages of increased TTHM levels from 2000-2015. The most significant increase in TTHMs in the West District was seen in Idaho (239%), and the lowest increase was in Montana (78%). The Midwest state with the most significant increase in TTHM was Kansas (87%), with all other states in the district having the lowest overall increases (below 22%). The South District's most significant increase was seen in Florida (294%), and the lowest increase was found in Oklahoma (29%). Lastly, the Northeast District had the most substantial increases overall, with the most significant increase in the seventh smallest state, Massachusetts (450%). Massachusetts has a population of approximately 6.5 million people in 10,555 square miles (27,340 km<sup>2</sup>), which consists of 25.7% water.



Recently the FDA banned TCS, but only from specific soap and hand sanitizing products; we want to look at the level of TTHM in drinking water after the ban went into effect in 2016. Antimicrobial TCS remains in many consumer products, including many popular toothpaste and mouth rinses. Consumers are exposed to these organochlorine-containing products, as evidenced by numerous studies showing detectable levels of TCS in skin, urine, and blood plasma ranging from 0.05 to 5.0  $\mu\text{M}$  concentrations (Wilburn et al., 2020). There remains controversy regarding whether TCS concentrations absorbed into the human body might induce adverse effects noted in lab studies. Considering these significant findings, incorporating this antimicrobial into readily available consumer products, not just in soap, needs to be re-evaluated, and the biological effects of its breakdown products and metabolites need to be investigated, especially in aquatic environments of our biosphere.

### Acknowledgements

We acknowledge the financial support from the Office of Research and Sponsored Programs (RSP) and the Title III Office under the supervision of Dr. Andrea Tyler of Tennessee State University. We also thank the United States water service offices for providing annual water safety reports.

### Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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## Abbreviations and Acronyms

EPA	Environmental Protection Agency
FDA	Food and Drug Administration
SDWIS/FED	Safe Drinking Water Information System/Federal Reporting Services
CCR	Consumer Compliance Report
CDC	Centers for Disease Control and Prevention
MWR	Major Water Report
EWG	Environmental Work Group
TCS	Triclosan
TTHM	Total Trihalomethanes