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A reduction in triclosan and triclocarban in water resource recovery facilities' influent, effluent, and biosolids following the U.S. Food and Drug Administration's 2013 proposed rulemaking on antibacterial products

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Additional Supporting Information may be found in the online version of this article.

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• Abstract

Pharmaceutical and personal care product compounds (PPCPs) comprise a large and diverse group of chemical compounds, including prescription and over-the-counter drugs and cleaning agents. Although PPCPs in the effluent and biosolids of water resource recovery facilities (WRRFs) are currently not regulated, public interest has led the Metropolitan Water Reclamation District of Greater Chicago to monitor for 11 PPCPs in the influent, effluent, and biosolids at its seven WRRFs. In 2016, the U.S. Food and Drug Administration (FDA) issued a final rule establishing that 19 specific ingredients, including triclosan and triclocarban, were no longer generally recognized as safe and effective, which prohibits companies from marketing soaps as antibacterial if they contain one or more of these ingredients. It was presumed that since the proposed rulemaking in 2013, manufacturers began to remove these active ingredients from their products. Annual monitoring of 11 PPCPs from 2012 to 2017 demonstrated a 71% decrease in triclosan and 72% decrease in triclocarban in per capita influent loading into seven WRRFs. There was a 70% decrease in triclosan and 80% decrease in triclocarban concentrations in biosolids. These declines suggest the FDA rule for the reduction in use of these compounds was effective and resulted in manufacturers removing these ingredients from their products.

• Practitioner points

- Reduction in triclosan and triclocarban per capita influent loading observed from 2012 to 2017.
- Reduction in triclosan and triclocarban biosolids loading observed from 2012 to 2017.
- 2016 FDA rulemaking on antimicrobial soaps was effective in removing triclosan and triclocarban from these products.
- Positive impact on quality of biosolids land applied to farmland.

• Key words

biosolids; effluent; influent; personal care products; pharmaceuticals; triclocarban; triclosan

Introduction

Pharmaceutical and personal care product compounds (PPCPs) generally refer to compounds found in any product used by individuals for health or cosmetic reasons or agribusiness to enhance the growth or health of livestock. These compounds comprise a large and diverse group of chemical substances, including prescription and over-the-counter drugs, veterinary drugs, fragrances, cosmetics, and cleaning agents. Pharmaceutical and personal care product compounds are continuously released into wastewater streams from domestic use and ultimately reach water resource recovery facilities (WRRFs) where they biodegrade, chemically degrade, partition to sludge solids, or remain soluble in the effluent (Luo et al., 2014; Onesios, Jim, & Bouwer, 2009; Zuehlke, Duennbier, Lesjean, Gnirss, & Buisson, 2006). There has been increasing attention given to the fate and transport of these compounds, including triclosan and triclocarban, in the environment from biosolids application to land (Boxall et al., 2012; Kinney et al., 2006; Prosser, Lissemore, Topp, & Sibley, 2014). Although PPCPs are not currently regulated in WRRFs' effluent or biosolids, continued public attention to trace concentrations of PPCPs in the environment led the Metropolitan Water Reclamation District of Greater Chicago (MWRDGC) to monitor for 11 select PPCPs in the influent, effluent, and biosolids at their seven WRRFs.

In 2016, the U.S. Food and Drug Administration (FDA) issued the final rule Safety and Effectiveness of Consumer Antiseptics; Topical Antimicrobial Drug Products for Over-the-Counter Human Use, 21 § CFR 310, which established that 19 specific ingredients, including triclosan and triclocarban, were no longer considered generally recognized as safe and effective (GRAS/GRAE) and were misbranded. This change in recognition by the FDA prohibits companies from marketing soaps or washes as antibacterial if they contain one or more of these 19 ingredients (Supporting information Table S1). It was presumed that manufacturers began to phase out the use of these active ingredients in their products since the FDA's proposed rulemaking in 2013. Monitoring data from 2012 to 2017 were analyzed to assess whether there was a change in triclosan and triclocarban concentrations in the MWRDGC's influent and biosolids from their seven WRRFs in response to a proposed change in the FDA's policy prohibiting companies from marketing soaps or washes as antibacterial. Policy changes such as this that affect the formulation of consumer products could have a significant and detectable effect of reducing the concentrations of trace organic compounds entering WRRFs.

Methods

Influent, effluent, and biosolids (digester draw or waste-activated sludge) samples were collected annually from the MWRDGC's seven WRRFs from 2012 to 2017. Influent and effluent loading values for 11 PPCPs were calculated using concentrations and daily flow on the day of sampling and then normalized on a per capita basis using 2010 census data for each facility's service area (Table 1). Normalizing data on a per capita basis allowed for the comparison of values across different size facilities. The James C. Kirie (Kirie), Lemont, and Terrence J. O'Brien (O'Brien) facilities send waste-activated sludge to the Stickney or the John E. Egan (Egan) facility for digestion. Only four of the seven water resource recovery facilities produce final biosolids: Calumet, Egan, Hanover Park, and Stickney. Nearly all of the MWRDGC's biosolids are ultimately beneficially reused through land application. The 11 PPCPs in the monitoring program were chosen because they are commonly used compounds, cover a range of different categories (e.g., anticonvulsant, antibiotic, antidepressant), and were evaluated in the U.S. Environmental Protection Agency (USEPA), 2009 Targeted National Sewage Sludge Survey (TNSSS) (Supporting information Table S2; USEPA, 2009).

A 24-hr composite sample, comprised of six sub-samples taken every 4 hr, was taken manually each year from each plant in either January or February. Generally, no more than two plants were sampled on the same day. Samples were collected at each facility's influent, effluent, and solids sampling point directly into one-gallon amber glass vials until full (no head space) and placed on ice in coolers for transport to the MWRDGC's Organic Compounds Analytical Laboratory. Empty vials were carried from the laboratory to the sampling site and returned unopened to the laboratory to monitor for contamination attributable to field handling and transportation procedures. Sample extractions were performed on whole samples without filtration prior to using the Oasis Disk (47 mm HLB) on an Automated Solid Phase Extractor (Horizon SPE-DEX 4790) using modified U.S. Environmental Protection Agency Method 1694 (USEPA, 2007). Analyses of compounds were performed with high-performance liquid chromatography with a triple quad mass spectrometer (Agilent 1200 HPLC and 6410B Triple Quad MS).

To ensure quality control of MWRDGC's PPCP analyses, precleaned and precertified bottles were purchased for sample use. Method blanks were prepared with the same analysis procedure as samples to ensure no contamination or interference. The method detection limit procedure in the Guidelines Establishing Test Procedures for the Analysis of Pollutants, 21 § CFR 136 (2017) was followed to extract and analyze seven replicates of low-level spikes and detection limits calculated following the recommended procedure. Final laboratory detection limits were adjusted by sample volume used for extraction or by dilution factor if the extract needed to be diluted. Laboratory control samples (i.e., known concentrations of target compounds spiked into reagent water), matrix spikes (i.e., known concentrations of target compounds spiked into a sample), and matrix spike duplicates (i.e., known concentrations of target compounds spiked into the same sample as matrix spikes) were also analyzed. Compound recoveries were monitored to ensure they met method requirements. Relative percent differences between matrix spikes and matrix spike duplicates were calculated. Surrogates (i.e., known concentrations of two surrogates spiked into all samples in order to calculate recoveries) were also calculated and monitored. A six- or seven-point calibration curve was conducted on the HPLC triple quad MS and relative standard deviations calculated by instrument software for all compounds to ensure proper calibration prior to analysis.

Percent solids for digester draw and waste-activated sludge samples were determined by weight after oven-drying the samples at 105°C for 24 hr. Percent solids ranged from 0.5% to 3.0%. Biosolids concentrations are reported on an oven-dried weight basis. Biological oxygen demand (BOD), suspended solids (SS), ammonia, and total phosphorus influent data for

Table 1. The Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities, populations serviced, treatment type, and description of biosolids samples

FACILITY NAME	AVERAGE FLOW[A] M^3/DAY (MGD)	TOTAL CAPACITY	POPULATION SERVICED[B]	TREATMENT TYPE	BIOSOLIDS SAMPLE
Stickney	2.6×10^6 (677)	5.5×10^6 (1,440)	2,160,235	Single stage nitrification; Anaerobic digestion	Digester draw
O'Brien	8.8×10^5 (232)	1.7×10^6 (450)	1,313,500	Single stage nitrification; Solids sent to Stickney	Waste-activated
Calumet	9.7×10^5 (256)	1.6×10^6 (430)	1,005,870	Conventional activated sludge; Anaerobic digestion	Digester draw
Kirie	1.6×10^5 (42)	1.6×10^6 (110)	264,667	Single stage/two stage nitrification; Solids sent to Egan	Waste-activated
Egan	9.1×10^4 (24)	1.9×10^5 (50)	160,735	Single stage nitrification; Anaerobic digestion	Digester draw
Hanover Park	3.6×10^4 (9.5)	8.3×10^4 (22)	56,532	Single stage nitrification; Anaerobic digestion	Digester draw
Lemont	9.8×10^3 (2.6)	1.5×10^4 (4.0)	21,113	Conventional activated sludge; Solids sent to Stickney	Waste-activated
Total	4.7×10^6 (1,243)	9.5×10^6 (2,506)	4,982,652[c]		

Notes. MGD: million gallons per day.

[a]2017 data.

[b]Based on 2010 Census data.

[c]The population equivalent for entire service area including domestic use and industrial discharge is approximately 10 million people.

corresponding dates and facilities were downloaded from the MWRDGC's Water Reclamation Plant Data published on its website (www.mwrd.org).

The resulting data set was small and not considered representative of the overall contaminant profile for each facility; however, statistical differences in triclosan and triclocarban concentrations that are not found for the other PPCPs would suggest a change in concentration did occur for these compounds. Due to this small sample size and lack of normal distribution in the annual influent, effluent, and biosolids data, significant differences were first determined by the Kruskal–Wallis rank sum test, which ranks the data and tests for differences in the locations of the distributions between groups. When the Kruskal–Wallis rank sum test was found to be significant ($p < 0.05$), the Dunn post hoc test for multiple comparisons was performed to determine significant differences between years ($p < 0.05$). All statistical analyses were conducted in the R software package v.3.4.1.

Results and Discussion

Despite a small data set from annual PPCP monitoring, evident trends in the data were observed for triclosan and triclocarban, but not for the other nine PPCPs analyzed. The mean per capita influent loading for triclosan decreased 71% from 2,811 μg day^{-1} person^{-1} in 2012 to 811 μg day^{-1} person^{-1} in 2017 (Figure 1). The mean per capita influent loading for triclocarban decreased 72% from 1,627 μg day^{-1} person^{-1} in 2012 to 455 μg day^{-1} person^{-1} in 2017 (Figure 2). No consistent decreases in mean per capita influent loading from 2012 to 2017 were observed for the other nine PPCPs (Table 2). Additionally, there were no significant differences from 2012

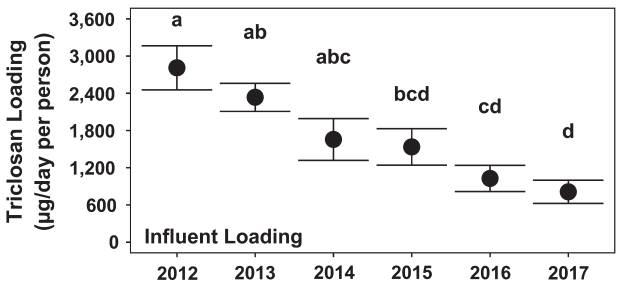

Figure 1. Decrease in mean ± *SE* per capita influent loading (µg day^{-1} person^{-1}) of triclosan in the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities. Different letters between years for the same compound indicate significant differences at $p < 0.05$ using Kruskal-Wallis rank sum test with post hoc Dunn test for multiple comparisons.

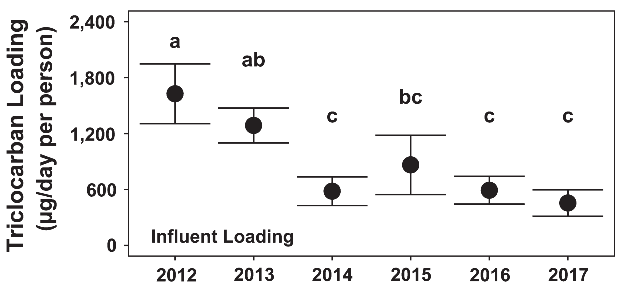

Figure 2. Decrease in mean ± *SE* per capita influent loading (µg day^{-1} person^{-1}) of triclocarban in the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities. Different letters between years for the same compound indicate significant differences at $p < 0.05$ using Kruskal-Wallis rank sum test with post hoc Dunn test for multiple comparisons.

to 2017 for BOD, SS, ammonia, or total phosphorus per capita influent concentrations and the values for these water quality analytes were in the range expected for typical WRRFs (Supporting information Table S3). The consistency of the other nine PPCPs and water quality analytes for the 8-year period helps to validate these observed reductions in triclosan and triclocarban.

Andrade et al. (2015) monitored the influent loading of select chemicals of concern six times annually over a 7 year period from 2005 to 2011 for a single WRRF and demonstrated a 42% decrease in brominated diphenyl ether (BDE)-47 and BDE-49 concentrations and a 47% decrease in triclocarban concentrations. These reductions in influent concentrations were attributed to these chemicals being phased out of manufacturing. The authors did not, however, observe any trends in triclosan over the same period. Krogh, Lyons, and Lowe (2017) found statistically significant decreases in triclosan concentrations from 2014 to 2016 at two WRRFs in Canada and a decrease in triclocarban concentrations of one of those two facilities. The concentrations of 10 other detected PPCPs at these facilities were stable over the same time period.

There were no consistent significant decreases in mean per capita effluent loading for any PPCP (Table 3). The 2012 effluent loading concentrations (non-normalized data) for triclocarban and triclosan for all seven plants ranged from 0.91 to 407 g/day with a mean of 117 and 0.74–158 g/day with a mean of 55 g/day, respectively. These values are comparable to reported effluent loading concentrations of 1.6–168 g/day for triclocarban and 0.91–76 g/day for triclosan from four WRRFs in Savannah, GA with flows ranging from 5.1×10^3 to 7.3×10^4 m^3/day (1.3–19 MGD) (Kumar, Priya, Peck, & Sajwan, 2010). Additionally, Heidler, Sapkota, and Halden (2006) reported an effluent loading of 127 g/day for triclocarban from a WRRF in the Mid-Atlantic region with a flow of 6.8×10^5 m^3/day (180 MGD).

Percent removal was calculated for the nine PPCPs other than triclosan and triclocarban, because they did not demonstrate a decrease in influent loading during the 8-year period. There were no additional studies conducted at these facilities to determine whether abiotic degradation, biodegradation, or sorption was the predominant mechanism for removal, so percent removal is considered inclusive of all processes.

Table 2. Mean ± *SE* per capita influent loading of pharmaceutical and personal care product compounds in the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities before and after the U.S. FDA's 2013 proposed rulemaking

	2012	2013	2014	2015	2016	2017
PPCPS	MG DAY^{-1} PERSON^{-1}					
Carbamazepine	108 ± 9.3a	137 ± 15a	187 ± 78a	128 ± 11a	103 ± 8.9a	204 ± 49a
Ciprofloxacin	2,258 ± 326a	1,921 ± 276ab	779 ± 138c	1,232 ± 247bc	1,372 ± 256abc	2,263 ± 524a
Codeine	52 ± 11a	107 ± 17a	89 ± 15a	104 ± 14a	111 ± 26a	126 ± 19a
Diphenhydramine	281 ± 61b	433 ± 49b	327 ± 51b	471 ± 53b	410 ± 56b	756 ± 51a
Fluoxetine	21 ± 2.2a	33 ± 4.7a	22 ± 3.7a	24 ± 4.2a	29 ± 4.2a	26 ± 3.0a
Gemfibrozil	613 ± 94a	685 ± 94a	567 ± 122a	633 ± 71a	529 ± 55a	428 ± 37a
Ibuprofen	5,369 ± 662a	6,620 ± 260a	6,128 ± 554a	6,494 ± 535a	6,016 ± 482a	6,121 ± 521a
Naproxen	5,161 ± 591a	6,209 ± 499a	6,603 ± 759a	6,949 ± 659a	6,224 ± 634a	6,113 ± 783a
Thiabendazole	13 ± 1.9a	26 ± 6.0a	18 ± 3.3a	15 ± 2.7a	18 ± 2.4a	20 ± 2.3a
Triclocarban	1,627 ± 320a	1,286 ± 187a	581 ± 154c	863 ± 318bc	592 ± 149c	455 ± 141c
Triclosan	2,811 ± 355a	2,335 ± 226ab	1,655 ± 337abc	1,536 ± 294bcd	1,027 ± 211 cd	811 ± 187d

Note. PPCPs: Pharmaceutical and personal care product compounds; Different letters between years for the same compound indicate significant differences at $p < 0.05$ using Kruskal–Wallis rank sum test with post hoc Dunn test for multiple comparisons.

Table 3. Mean ± *SE* per capita effluent loading of pharmaceutical and personal care product compounds from the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities before and after the U.S. FDA's 2013 proposed rulemaking

	2012	2013	2014	2015	2016	2017
PPCPS	MG DAY^{-1} PERSON^{-1}					
Carbamazepine	97 ± 7.6a	148 ± 16a	120 ± 16a	118 ± 14a	104 ± 6.8a	124 ± 14a
Ciprofloxacin	900 ± 108a	501 ± 96b	304 ± 40b	350 ± 38b	409 ± 60b	330 ± 82b
Codeine	53 ± 10b	102 ± 15a	95 ± 17ab	93 ± 12ab	62 ± 14b	126 ± 19a
Diphenhydramine	167 ± 12a	246 ± 17a	213 ± 38a	201 ± 30a	166 ± 32a	234 ± 28a
Fluoxetine	13 ± 1.3a	21 ± 4.4a	15 ± 2.1a	14 ± 0.89a	17 ± 2.1a	15 ± 1.4a
Gemfibrozil	251 ± 70a	297 ± 56a	383 ± 64a	321 ± 49a	183 ± 42a	208 ± 60a
Ibuprofen	179 ± 87a	113 ± 39a	169 ± 103a	75 ± 36a	122 ± 95a	28 ± 12a
Naproxen	178 ± 73a	159 ± 71a	778 ± 459a	151 ± 76a	153 ± 76a	147 ± 69a
Thiabendazole	12 ± 2.0a	28 ± 10a	14 ± 2.6a	13 ± 2.2a	15 ± 2.1a	18 ± 2.3a
Triclocarban	140 ± 48a	97 ± 21a	87 ± 19a	67 ± 23a	42 ± 13a	51 ± 15a
Triclosan	73 ± 17a	83 ± 24a	123 ± 61a	65 ± 15a	66 ± 15a	45 ± 12a

Note. Different letters between years for the same compound indicate significant differences at $p < 0.05$ using Kruskal–Wallis rank sum test with post hoc Dunn test for multiple comparisons.

Table 4. Percent removal and log Kow values for nine pharmaceutical and personal care product compounds from the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities

COMPOUND	PERCENT REMOVAL[a]	LOG KOW
Gemfibrozil	52	4.77
Fluoxetine	39	4.05
Ibuprofen	98	3.97
Naproxen	96	3.18
Diphenhydramine	54	3.27
Thiabendazole	9.0	2.47
Carbamazepine	18	2.45
Codeine	10	1.19
Ciprofloxacin[b]	77	0.28

[a]Percent removal determined as difference between mean influent and mean effluent loading for the 8-year period.

[b]2013–2017 data only.

Percent removal was determined as the percent decrease between the mean influent and effluent concentrations for the 8-year period and ranged from 9% to 98% for the nine PPCPs (Table 4).

One of the key mechanisms for removal of PPCPs from WRRFs is by sorption to biosolids (Onesios et al., 2009). The sorption of PPCPs to biosolids is dependent on lipophilicity, expressed as log Kow. Generally, a log Kow <2.5 results in low sorption potential, log Kow >2.5 and <4.0 yields medium sorption potential, and log Kow >4.0 promotes high sorption potential (Jones-Lepp & Stevens, 2007). The PPCPs with the greatest removal, ibuprofen and naproxen, had log Kow values <4.0, and the PPCPs with log Kow values >4.0, gemfibrozil and fluoxetine, had removals of only 52% and 39%, respectively (Table 4). Ciprofloxacin also had a notably high removal rate of 77% (2013–2017), but has a low log Kow value of 0.28. These results suggest a likely mix of sorption and degradation processes contributing to the removal of PPCPs from these facilities. Overall, the removal of these PPCPs during this 8-year period was in the range of previously reported values (Kumar et al., 2010; Rosal et al., 2010; Xia, Bhandari, Das, & Pillar, 2006).

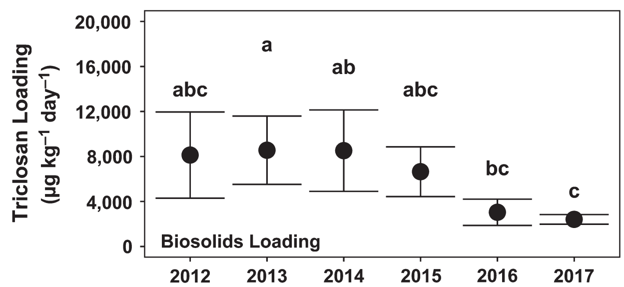

Figure 3. Decrease in mean ± *SE* concentration of triclosan in biosolids from the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities. Different letters between years for the same compound indicate significant differences at $p < 0.05$ using Kruskal-Wallis rank sum test with post hoc Dunn test for multiple comparisons.

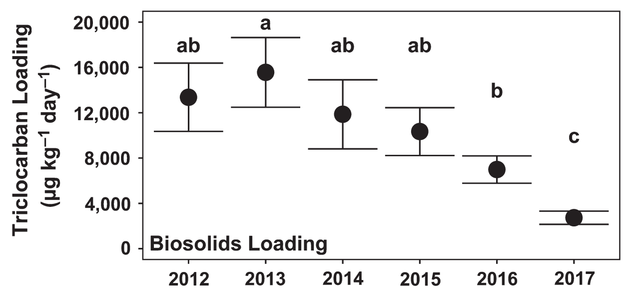

Figure 4. Decrease in mean ± *SE* concentration of triclocarban in biosolids from the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities. Different letters between years for the same compound indicate significant differences at $p < 0.05$ using Kruskal-Wallis rank sum test with post hoc Dunn test for multiple comparisons.

Table 5. Mean ± *SE* concentrations of pharmaceutical and personal product compounds in the biosolids generated at the Metropolitan Water Reclamation District of Greater Chicago's seven water resource recovery facilities before and after the U.S. FDA's 2013 proposed rulemaking

	2012	2013	2014	2015	2016	2017
PPCPS	MG KG^{-1} DAY^{-1}					
Carbamazepine	62 ± 14a	81 ± 14a	89 ± 21a	79 ± 21a	64 ± 21a	48 ± 8.0a
Ciprofloxacin	20,834 ± 3,844a	17,834 ± 4,314a	10,851 ± 2,752a	14,470 ± 3,154a	12,035 ± 1,753a	14,918 ± 4,002a
Codeine	16 ± 5.5a	53 ± 18a	33 ± 18a	56 ± 20a	46 ± 16a	73 ± 19a
Diphenhydramine	564 ± 104a	882 ± 140a	611 ± 132a	676 ± 123a	643 ± 93a	674 ± 91a
Fluoxetine	131 ± 28a	180 ± 47a	160 ± 37a	158 ± 39a	129 ± 19a	88 ± 20a
Gemfibrozil	161 ± 50ab	284 ± 64a	182 ± 58ab	238 ± 48a	154 ± 28ab	73 ± 8.9b
Ibuprofen	766 ± 231a	1,418 ± 373a	1,001 ± 281a	1,128 ± 360a	960 ± 250a	431 ± 69a
Naproxen	104 ± 54a	173 ± 70a	223 ± 89a	267 ± 107a	116 ± 51a	169 ± 81a
Thiabendazole	41 ± 20a	125 ± 77a	36 ± 9.7a	39 ± 8.5a	75 ± 52a	29 ± 3.3a
Triclocarban	13,362 ± 3,011ab	15,557 ± 3,072a	11,859 ± 3,049ab	10,335 ± 2,112ab	6,985 ± 1,208b	2,730 ± 584c
Triclosan	8,119 ± 3,831abc	8,552 ± 3,038a	8,516 ± 3,619ab	6,643 ± 2,211abc	3,034 ± 1,170bc	2,408 ± 429c

Note. PPCPs: Pharmaceutical and personal care product compounds; Different letters between years for the same compound indicate significant differences at $p < 0.05$ using Kruskal–Wallis rank sum test with post hoc Dunn test for multiple comparisons.

There were observed trends in PPCP concentrations in biosolids that were also unique to triclosan and triclocarban. From 2012 to 2017, the mean triclosan concentration in MWRDGC's biosolids decreased by 70% from 8,119 to 2,408 μg/kg (Figure 3) and the mean triclocarban concentration decreased by 80% from 13,363 to 2,730 μg/kg (Figure 4). There were no consistent trends in the mean concentrations in biosolids for the other nine PPCPs over the same time period (Table 5). The EPA's 2009 TNSSS reported mean triclosan and triclocarban concentrations in biosolids at 16,097 and 39,433 μg/kg, respectively (USEPA, 2009; Supporting information Table S2). The mean concentrations of 2,408 μg/kg for triclosan and 2,703 μg/kg for triclocarban in MWRDGC's biosolids in 2017 were 85% and 93% lower, respectively, than concentrations reported in the 2009 TNSSS. This suggests that a new national survey of biosolids may find significantly reduced concentrations of these two PPCPs.

The nearly year-to-year decrease in triclocarban and triclosan loading in influent from 2012 to 2017 in the MWRDGC's seven facilities, in combination with a lack in year-to-year changes in the loading for nine other PPCPs and water quality parameters, suggests that triclosan and triclocarban loading to WRRFs decreased in response to the FDA's proposed rulemaking in 2013. The reduction in triclosan and triclocarban loading in the influent resulted in a corresponding reduction in these compounds in the biosolids generated at these WRRFs. Reductions in the concentrations of trace organic compounds help to improve the quality of biosolids, which is an ongoing goal for WRRFs recovering resources through the land application of biosolids.

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