ORGANIC WASTEWATER CONTAMINANTS IN THE UPPER CHATTAHOOCHEE RIVER BASIN, GEORGIA, 1999–2002

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Abstract. The U.S. Geological Survey recently developed a laboratory method designed to analyze organic wastewater contaminants (OWCs) typically found in domestic and industrial wastewater. As part of five different studies conducted from 1999 through 2002, 74 water samples were collected at 26 sites in the upper Chattahoochee River Basin and analyzed for OWCs. In general, the number of OWCs detected and the concentrations of OWCs measured at various site types decreased from effluent samples, to stream samples collected during wet-weather conditions, to stream samples collected during baseflow conditions, to drinking-water samples.

Although few of the OWCs measured have drinking-water standards or other human or ecological health criteria, there were 3 exceedences of the maximum contaminant level for the insecticide diazinon and 12 exceedences of aquatic-life criteria for the insecticides diazinon, carbaryl, and chlorpyrifos. The total concentration of 13 potential endocrine disruptors measured as part of the OWC method exceeded 1 microgram per liter in most samples collected from treated effluent and from tributary streams during wet-weather conditions. Little is known, however, about the potential health effects to humans or aquatic organisms exposed to low levels of most of these chemicals, or more importantly, to mixtures of these chemicals.

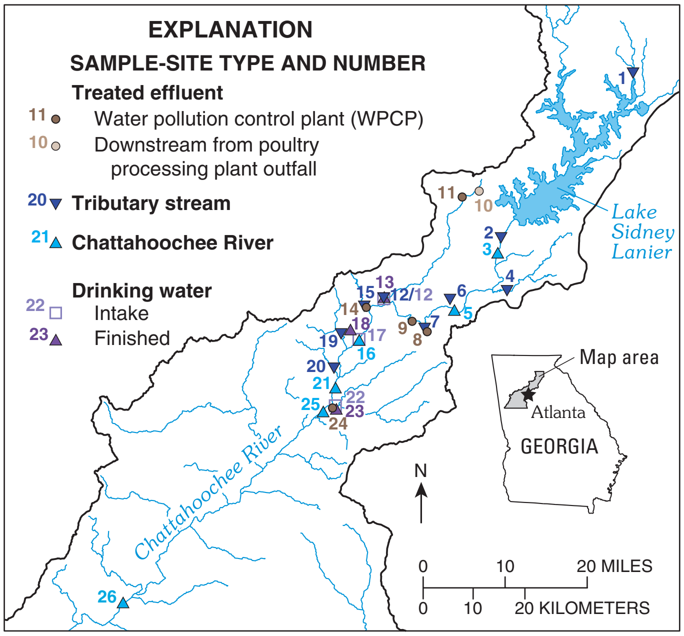
Several patterns in the number and relative concentrations of OWCs detected among the various site types indicate the potential to use OWCs to identify the likely presence or absence of wastewater in specific samples or sites at various flow conditions. For example, detection frequencies and relative concentrations of some OWCs, such as several plasticizers and (or) fire retardants and detergent metabolites, at various sites and flow conditions may help locate sources of wastewater to streams upstream from sampling locations.

INTRODUCTION

Some chemicals used daily in homes, industry, and agriculture enter the aquatic environment in treated and untreated wastewater effluent, stormwater runoff, and ground-water seepage. The U.S. Geological Survey (USGS) recently developed a laboratory method designed to analyze compounds typically found in domestic and industrial wastewater. Organic wastewater contaminants (OWCs) measured include nonionic surfactant compounds (including detergent metabolites) that are persistent indicators of wastewater and compounds representative of food additives, antioxidants, fire retardants, plasticizers, solvents, disinfectants, animal steroids, polycyclic aromatic hydrocarbons (PAH), and high-use domestic pesticides (Brown and others, 1999; Barber and others, 2000). Most compounds analyzed have anthropogenic sources, although a few, such as cholesterol, coprostanol, and stigmastanol, also occur naturally. Although each of the OWCs analyzed is used extensively, little information is available about the occurrence of many of these compounds in the environment. Some are indicators of contamination sources, such as human waste, and some have human or environmental health implications.

The Chattahoochee River is one of Georgia's most utilized water resources—supplying drinking water to a large percentage of the Metropolitan Atlanta population and serving as a receiving waterbody for treated wastewater as well as untreated urban runoff. As part of five separate studies (Fig. 1), 74 OWC samples were collected from May 1999 to September 2002 at 26 sites in the upper Chattahoochee River Basin. Criteria used for site selection varied among the five studies, but the 26 sites represent a variety of predominant land uses, watershed sizes, and site types and are located upstream, within, and downstream from Metropolitan Atlanta (Fig. 1).

The OWC data presented in this report can be used as a chemical method of bacterial or fecal source tracking (BST). For the subset of sites sampled as part of the USGS and National Park Service study of microbial contamination in streams within the Chattahoochee River National Recreation Area (study 2 in Fig. 1), fecal-indicator bacteria data, molecular BST data (ribotypes of *Escherichia coli* [*E. coli*]; Hartel and others, 2001), and biochemical BST data (F-specific and somatic coliphage



Site number	Station name	Studies in which sites were sampled 1	2	3	4	5
1	West Fork Little River	x			x	
2	James Creek		x			
3	Chattahoochee River at Settles Bridge		x			
4	Suwanee Creek		x			
5	Chattahoochee River near Norcross					x
6	Johns Creek		x			
7	Crooked Creek		x			
8	Crooked Creek WPCP			x		
9	Johns Creek WPCP			x		
10	Kelly Mill Branch Tributary			x		
11	City of Cummings WPCP			x		
12	Big Creek below Water Works Intakes		x	x		
13	City of Roswell Water Treatment Plant			x		
14	Big Creek WPCP			x		
15	Willeo Creek		x			
16	Chattahoochee River at Johnson Ferry Road		x			
17	Cobb County Water Intake			x		
18	Cobb County Water Treatment Plant			x		
19	Sope Creek		x			
20	Rottenwood Creek		x			
21	Chattahoochee River at Atlanta		x			
22	Atlanta Water Works Intake			x		
23	Atlanta Water Works - Chattahoochee Water Treatment Plant			x		
24	R.M. Clayton WPCP					x
25	Chattahoochee River at State Route 280		x			x
26	Chattahoochee River near Whitesburg	x				x

Base modified from U.S. Geological Survey, variously scaled digital data

Figure 1. Location of sites sampled for organic wastewater contaminants, upper Chattahoochee River Basin, 1999–2002 (studies in which sites were sampled: 1, Kolpin and others, 2002; 2, Gregory and Frick, 2001; 3, Henderson and others, 2001, and Frick and others, 2001; 4, USGS Toxic Substances Hydrology Program unpublished emerging contaminant study in agriculural areas; and 5, Furlong and others, in press).

and *Clostridium perfringens*) were collected concurrently with the OWC data. Most BST methods are still considered experimental; however, confidence in results is higher when more than one BST method indicates similar locations of fecal contamination within a watershed (Sinton and others, 1998; and Scott and others, 2002).

METHODS

The sampling methods and quality assurance protocols are the same as those described in Kolpin and others (2002), with two exceptions. The first exception is that although integrated samples were collected when possible, grab samples were collected at water pollution control plants (WPCP) (Fig. 1: sites 8, 9, 11, 14, and 24), water treatment plants (WTP) (sites 13, 18, and 23), downstream from a poultry processing plant outfall (site 10), and at one tributary stream during wet-weather conditions (site 2). The second exception is that to reduce the likelihood of reporting a false positive detection, environmental concentrations within three times the values rather than twice the values observed in set blanks were reported as less than the reporting limit (RL). Brown and others (1999) and Barber and others (2000) described laboratory protocols for the OWC analytical method.

The specific compounds analyzed and the RLs varied among samples reported in this paper (Table 1) because of method refinement during the 4 years that samples were collected and analyzed and because of matrix interference. Figures 2, 3, and 4 only include compounds that were analyzed in 56 or more samples. Table 1 includes four additional compounds that were analyzed in only 17 samples.

RESULTS

From 1 to 22 OWCs were detected in 92 percent of the samples collected from the Chattahoochee River Basin (Fig. 2*A*). Five surface-water samples and one finished drinking-water sample collected during base-flow conditions in the midst of a multiyear drought had no OWCs detected. More than 92 percent of the detections of OWCs were at very low concentrations (less than 1 micrograms per liter [µg/L]); however, mixtures of chemicals were common and total concentrations often exceeded 1 µg/L. The median number of OWCs detected at various site types decreased from 15–16 in treated effluent, to 13–15 in stream samples collected during wet-weather conditions, to 2–6 in stream samples and samples from WTP intakes collected during base-flow conditions, to 2 in finished drinking water (Fig. 2 and Table 1). The median total concentration of OWCs detected at various site types decreased from 31 µg/L downstream from a poultry processing plant, to 8.5 µg/L in treated effluent from WPCP, to 4.1 µg/L in tributary stream samples collected during wet-weather conditions, to 1.7 µg/L in Chattahoochee River samples collected during wet-weather conditions, to 0.08–0.9 µg/L in stream samples and samples collected at WTP intakes during baseflow conditions, to 0.4 µg/L in finished drinking water (Fig. 2 and Table 1). The median number of OWCs detected and the median total concentration of OWCs detected by site type are likely to be underestimates since not all compounds were measured at all sites and some RLs in a few samples were larger than those listed in Table 1.

In 12 of 13 sites that were sampled during baseflow and wet-weather conditions and in findings from several streams sampled in the Metropolitan area of Kansas City, Missouri (Wilkison and others, 2002), a higher number of OWCs generally were detected in wet-weather samples than in baseflow samples. Concentrations of OWCs in wet-weather conditions also were often higher. Some possible explanations of why more OWCs at higher concentrations were detected in streams during wet-weather conditions include: (1) wastewater reaching streams from areas that are not typically sources during baseflow; (2) nonpoint sources of some OWCs that are unrelated to wastewater; (3) sanitary sewers that have overflow problems that may or may not occur during baseflow conditions; (4) increased contribution of septic tank leachate to streamflow through increased near-stream source area; and (5) wastewater compounds, many of which have high affinities for sediments, can be re-suspended from the disturbance of bottom sediments (Wilkison and others, 2002). Although the organic compounds measured are often detected in wastewater, these compounds have multiple uses and the predominant uses of some of these compounds have changed through time.

Few of the OWCs measured have drinking-water standards or other human or ecological health criteria (Kolpin and others, 2002). Samples collected from tributary streams during wet-weather conditions contained 3 exceedences of a maximum contaminant level (for the insecticide diazinon) and 12 exceedences of aquatic-life criteria (for the insecticides diazinon, carbaryl, and chlorpyrifos; Table 1). No other exceedences of available standards or criteria were observed.

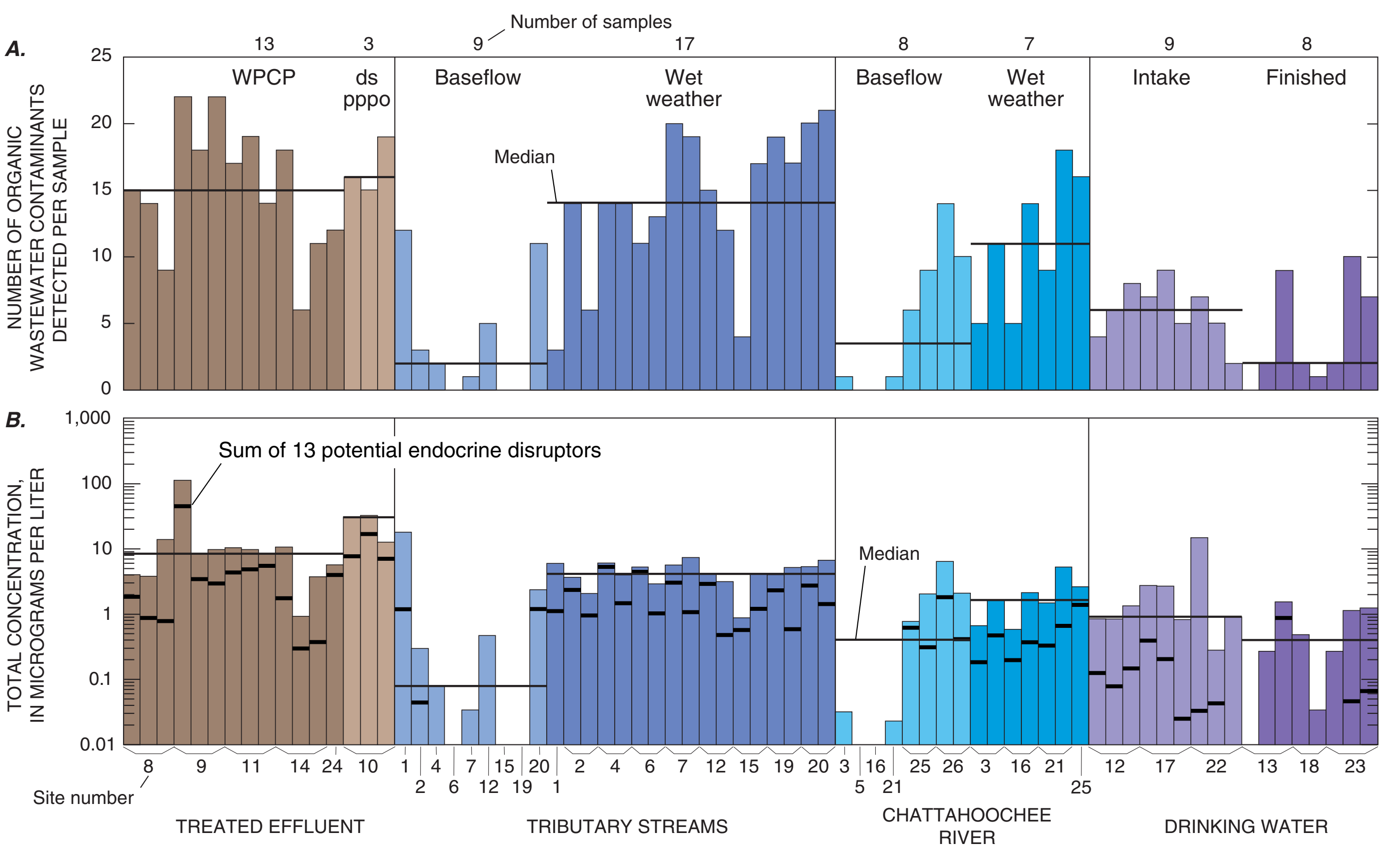


Figure 2. Summary of organic wastewater contaminants (OWCs) detected per sample for *(A)* number of compounds detected and *(B)* total concentration (summation of detections of 43 compounds; absence of bar shows no OWCs detected; WPCP, water pollution control plant; ds pppo, downstream from poultry processing plant outfall).

Table 1. Summary of organic wastewater compounds (OWCs) detected[/a], upper Chattahoochee River Basin, 1999–2002

[CASRN, Chemical Abstracts Service Registry Number; N, number of samples; RL, reporting limit; µg/L, microgram per liter; Max, maximum concentration; %, percent; WPCP, water pollution control plant; ds pppo, downstream from a poultry processing plant outfall; E, compound detected, but concentration estimated; na, not analyzed; LC_{50}, lethal concentration with 50 percent mortality; Chemicals that are potential endocrine disruptors are in bold; Percent detections in bold indicate which site type had the maximum concentration measured for a chemical]

Chemical	CASRN	N	RL (µg/L)	Max (µg/L)	Percent detections, by site type (number of analyses are listed in column heading; when more than one sample in a site type was not analyzed for a chemical, then the number of detections/number of analyses is listed instead of percent detections)							
					Treated effluent		Tributary stream		Chattahoochee River		Drinking water	
					WPCP, % (13)	ds pppo, % (3)	Base-flow, % (9)	Wet weather, % (17)	Base-flow, % (8)	Wet weather, % (7)	Intake, % (9)	Finished, % (8)
Nonprescription drugs - caffeine & nicotine metabolites												
caffeine	58-08-2	74	0.08[/b]	1.2	**69**	100	67	94	75	100	100	38
cotinine	486-56-6	17	0.04[/b]	E 0.2	**1/1**	na	na	2/9	2/4	3/3	na	na
Insect repellent												
N,N-diethyl-meta-toluamide (DEET)	134-62-3	17	0.04[/b]	0.12	0/1	na	na	7/9	2/4	**3/3**	na	na
Plasticizers and/or fire retardants												
tri(2-chloroethyl)phosphate	115-96-8	74	0.04[/b]	0.52	**100**	67	33	82	50	57	56	75
phthalic anhydride	85-44-9	69	0.5[/d]	E 1.6	75[/c]	0	11	**56**[/c]	0/5	14	11	0
tris(2-butoxyethyl) phosphate	78-51-3	74	0.07[/b]	38	**38**	100	22	82	13	43	33	38
tributylphosphate	126-73-8	61	0.04[/d]	1.9	**100**	0	0[/c]	7/9	43[/c]	0/4	0	25
bisphenol A	80-05-7	74	0.09[/b]	0.48	23	**100**	11	59	38	29	0	0
triphenyl phosphate	115-86-6	74	0.1[/b]	0.11	8	0	0	**41**	38	43	0	0
bis(2-ethylhexyl)phthalate	117-81-7	69	1.6[/d]	14	17[/c]	33	11	0[/c]	0/5	0	**11**	0
tri(dichlorisopropyl)phosphate	13674-87-8	74	0.1[/b]	E 0.23	**8**	0	0	0	25	0	0	0
bis(2-ethylhexyl) adipate	103-23-1	69	1.4[/b]	E 2	0[/c]	0	**11**	0[/c]	0/5	0	0	0
Steroids												
cholesterol	57-88-5	74	1[/b]	E 14	**85**	100	11	94	38	86	44	13
3*beta*-coprostanol	360-68-9	72	0.6[/b]	E 8.7	**77**	67	25[/c]	18	0[/c]	57	0	13
beta-stigmastanol [stigmastanol]	19466-47-8	17	2	E 2.1	0/1	na	na	**1/9**	0/4	0/3	na	na
Disinfectants												
triclosan	3380-34-5	74	0.04[/b]	2	**100**	100	22	82	38	57	11	38
phenol	108-95-2	74	0.4[/d]	7.9	23	**67**	0	0	25	0	33	0
para-Cresol (4-methyl phenol)	106-44-5	74	0.03[/b]	8.7	69	**67**	0	47	0	0	0	0
Polycyclic aromatic hydrocarbons (PAHs)												
fluoranthene	206-44-0	74	0.03[/d]	0.66	15	67	22	**76**	13	100	67	38
pyrene	129-00-0	74	0.03[/b]	0.45	15	67	22	**76**	13	100	67	38
phenanthrene	85-01-8	74	0.06[/b]	0.34	15	**33**	0	47	0	57	11	13
benzo(a)pyrene	50-32-8	74	0.05[/b]	0.18	8	33	0	**35**	0	57	22	13
anthracene	120-12-7	74	0.05[/d]	0.05	0	**33**	0	47	0	14	0	0
2,6-dimethylnapthalene	581-42-0	61	0.09[/b]	0.51	8	**33**	0[/c]	1/9	0[/c]	0/4	0	0
naphthalene	91-20-3	74	0.03[/b]	0.04	0	33	0	**12**	0	0	0	0
Insecticides												
diazinon	333-41-5	74	0.03[/b]	0.86	54	0	11	**94**[/e]	13	71	44	0
carbaryl	63-25-2	74	0.06[/b]	E 3.5	46	0	0	**88**[/f]	0	100	56	0
chlorpyrifos	2921-88-2	74	0.02[/b]	0.22	0	0	0	**29**[/g]	0	14	0	0
cis-chlordane	5103-71-9	69	0.04	E 0.02	8[/c]	0	**11**	0[/c]	0/5	0	0	0
lindane	58-89-9	69	0.05	E 0.04	17[/c]	0	0	0[/c]	0/5	0	0	0

Table 1. Summary of organic wastewater compounds (OWCs) detected[/a], upper Chattahoochee River Basin, 1999–2002—Continued

[CASRN, Chemical Abstracts Service Registry Number; N, number of samples; RL, reporting limit; µg/L, microgram per liter; Max, maximum concentration; %, percent; WPCP, water pollution control plant; ds pppo, downstream from a poultry processing plant outfall; E, compound detected, but concentration estimated; na, not analyzed; LC_{50}, lethal concentration with 50 percent mortality; Chemicals that are potential endocrine disruptors are in bold; Percent detections in bold indicate which site type had the maximum concentration measured for a chemical]

Chemical	CASRN	N	RL (µg/L)	Max (µg/L)	Percent detections, by site type (number of analyses are listed in column heading; when more than one sample in a site type was not analyzed for a chemical, then the number of detections/number of analyses is listed instead of percent detections)							
					Treated effluent		Tributary stream		Chattahoochee River		Drinking water	
					WPCP, % (13)	ds pppo, % (3)	Base-flow, % (9)	Wet weather, % (17)	Base-flow, % (8)	Wet weather, % (7)	Intake, % (9)	Finished, % (8)
Detergent metabolites												
nonylphenol, monoethoxylate - (total NPEO1)	na	74	0.08[/b]	E 20	**69**	100	22	29	13	14	11	0
***para*-nonylphenol (total, NP)**	84852-15-3	72	0.5[/b]	E 11	**46**	100	13[/c]	47	0[/c]	14	11	13
nonylphenol, diethoxylate - (total NPEO2)	26027-38-3	74	1[/d]	E 8.7	**69**	100	22	29	25	14	0	0
octylphenol, monoethoxylate (OPEO1)	26636-32-8	74	0.1[/d]	E 1.3	**31**	67	22	29	13	0	0	0
octylphenol, diethoxylate - (OPEO2)	26636-32-8	74	0.2[/b]	E 0.29	**23**	33	11	18	38	0	0	0
Fumigant												
1,4-dichlorobenzene	106-46-7	74	0.03[/b]	0.36	**85**	0	0	6	13	29	0	0
Antioxidants												
2,6-di-*tert*-butylphenol (DTBP)	128-39-2	69	0.09	E 0.22	50[/c]	0	0	0[/c]	0/5	0	0	38
butylated hydroxy toluene (BHT)	128-37-0	69	0.08	E 0.15	50[/c]	0	0	0[/c]	0/5	0	0	0
3-*tert*-butyl-4-hydroxy anisole (BHA)	25013-16-5	74	0.1[/d]	E 0.74	**38**	0	0	0	0	0	0	0
5-methyl-1H-benzotriazle	136-85-6	17	0.1[/b]	E 1.1	**1/1**	na	na	0/9	0/4	0/3	na	na
2,6-di-*tert*-butyl-1,4-benzoquinone (DTBB)	719-22-2	69	0.07[/d]	0.33	25[/c]	0	0	0[/c]	0/5	14	0	0
Solvents												
tetrachloroethylene	127-18-4	74	0.03[/b]	0.53	15	**100**	0	6	0	0	0	13
2(2-butoxyethoxy) ethyl acetate	124-17-4	56	0.06[/d]	0.30	0[/c]	0	0[/c]	2/8	0/4	**1/4**	0	0
Flavors and fragrances												
acetophenone	98-86-2	74	0.1[/h]	0.38	0	**67**	0	24	0	14	0	0
benzaldehyde	100-52-7	56	0.1[/b]	0.17	33[/c]	0	0[/c]	0/8	0/4	0/4	0	13
Prescription drug — analgesic												
codeine	76-57-3	69	0.1	0.55	25[/c]	0	0	0[/c]	0/5	0	0	0
Number of OWC compounds detected/ total number of OWCs analyzed					38/46	24/42	19/42	33/46	20/46	26/46	16/42	15/42
Median number of OWCs detected per sample					15	16	2	15	3.5	13	6	2
Median total concentration of OWCs detected per sample					8.5	31	0.08	4.1	0.4	1.7	0.9	0.4

[/a] The following five compounds were analyzed in at least 17 samples, but were not detected (RL, in µg/L): 1,2-dichlorobenzene (0.03); 1,3-dichlorobenzene (0.03); dieldrin (0.08); methyl parathion (0.06); diethyl phthalate (DEP) (0.25).

[/b] Predominant reporting limit is listed, two RLs in data set.

[/c] Percentage is based on one less sample analyzed than listed in parenthesis in heading for column.

[/d] Predominant reporting limit is listed, three RLs in data set.

[/c] Diazinon: 3 exceedences of maximum contaminant level (MCL) of 0.6 µg/L and 4 exceedences of LC_{50} of 0.56 µg/L.

[/f] Carbaryl: 7 exceedences of LC_{50} of 0.4 µg/L.

[/g] Chlorpyrifos: 1 exceedence of LC_{50} of 0.1 µg/L.

[/h] Predominant reporting limit is listed, four RLs in data set.

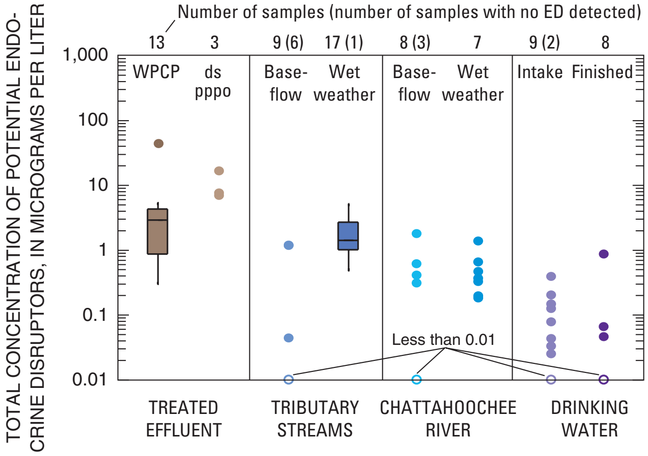


Figure 3. Total concentration of potential endocrine disruptors (ED), by site type (WPCP, water pollution control plant; ds pppo, downstream from poultry processing plant outfall).

Thirteen measured compounds are known or suspected to exhibit at least weak hormonal activity, with the potential to disrupt endocrine function (potential endocrine disruptors; Table 1, Figs. 2*B* and 3); however, little is known about the potential health effects to humans or aquatic organisms exposed to the low levels of most of these chemicals or the mixtures measured in this study. The total concentration of potential endocrine disruptors was highest in samples collected from treated effluent and from streams during wet-weather conditions (Figs. 2*B* and 3). No potential endocrine disruptors were detected in six of nine baseflow samples collected from tributary streams and in the four baseflow samples collected from the Chattahoochee River upstream from major wastewater outfalls at and near site 24 (Figs. 2*B* and 3). The baseflow samples with the most number and highest concentrations of potential endocrine disruptors and OWCs detected include the most urbanized tributary stream sampled (site 20) and the Chattahoochee River (sites 25 and 26) downstream from where most treated effluent from Metropolitan Atlanta is discharged (Fig. 2*B*). It is not clear what the sources of OWCs are at sites 1 and 2, which are watersheds with a large amount of poultry production and a less developed rural to suburban watershed, respectively.

Several patterns in the number and relative concentrations of OWCs detected among the various site types (Table 1) indicate the potential to use OWCs to identify the likely presence or absence of wastewater in specific samples or sites at various flow conditions. For example, the higher frequency of detection and concentrations of several PAHs and insecticides in samples collected from tributary streams during wet-weather conditions compared to treated effluent, indicates that the primary source(s) of specific PAHs and insecticides upstream from these sampling sites are probably nonpoint-source runoff rather than treated wastewater. Sinton and others (1998) stated that although PAHs occur in sewage, they are associated primarily with urban stormwater runoff, and their principal value is in distinguishing between stormwater and sewage contaminant sources. In contrast, detection frequencies and relative concentrations of some OWCs, such as several plasticizers and (or) fire retardants and detergent metabolites, at various sites and flow conditions may help locate sources of wastewater to streams upstream from sampling locations. For the subset of samples collected as part of one baseflow and two wet-weather synoptic sampling of 12 sites (study 2 in Fig. 1), all three methods of bacterial source tracking (chemical, molecular, and biochemical) indicate that fecal contamination is present in most if not all sites sampled during wet-weather conditions and in Rottenwood Creek and potentially a few other sites during baseflow conditions. For example, total concentrations of OWCs and *E. coli* concentrations were typically an order of magnitude higher in samples collected during wet weather conditions than samples collected during baseflow conditions and tended to be higher in tributary streams than in the Chattahoochee River (Fig. 4). No OWC data were available for untreated wastewater in the Chattahoochee River Basin, which is an important sample type for future studies to track potential wastewater sources.

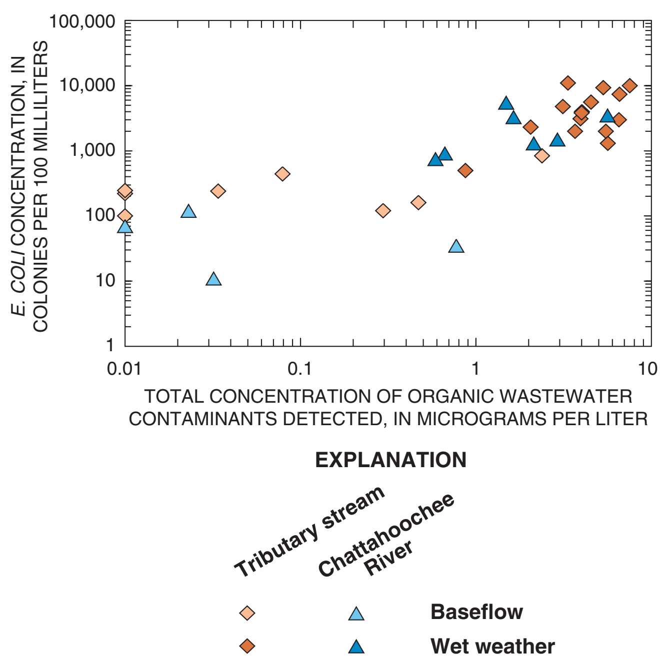


Figure 4. Relation between the total concentration of organic wastewater contaminants detected and *E. coli* concentrations (only includes sites sampled as part of study 2 in Figure 1).

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