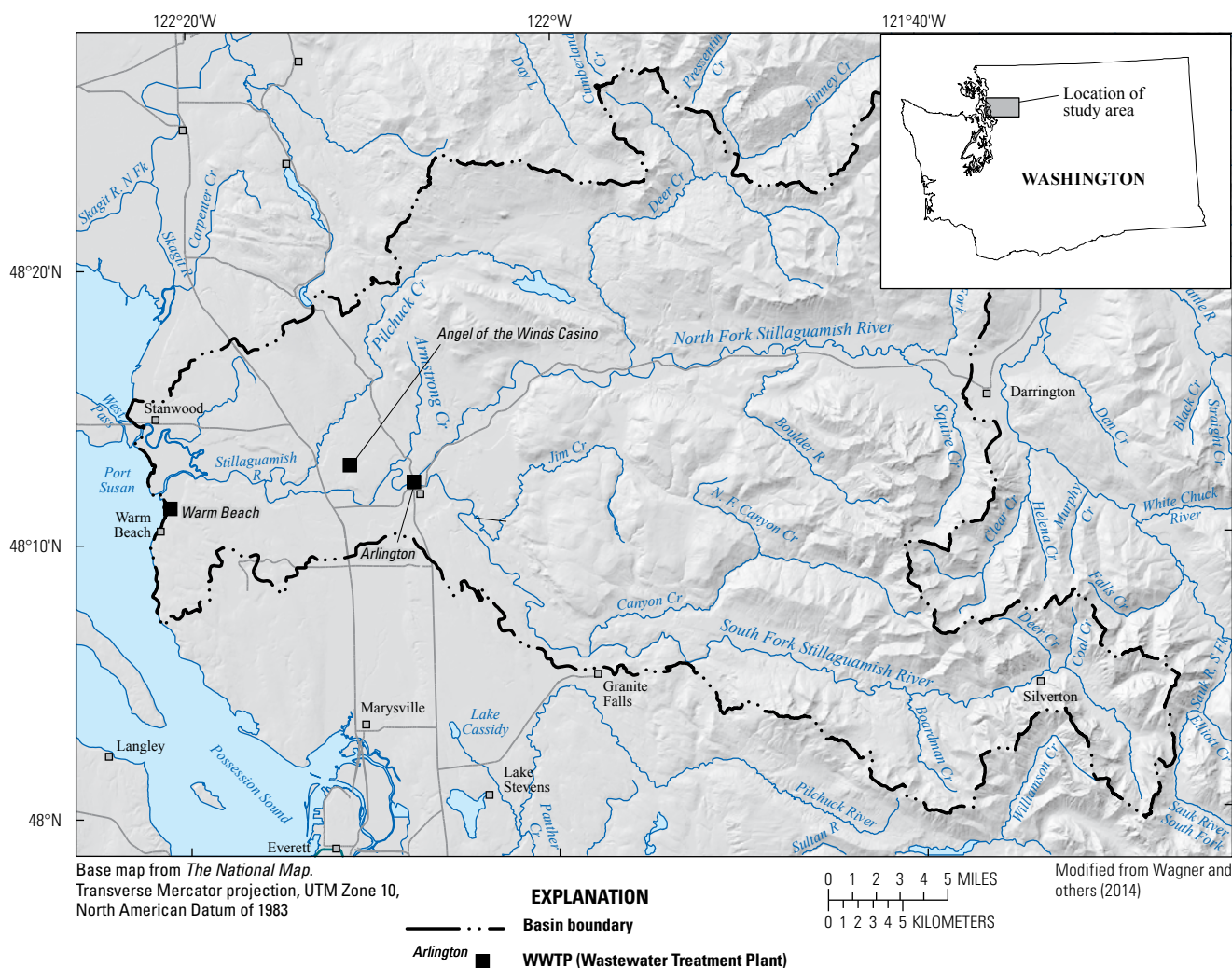


# Contaminant Removal by Wastewater Treatment Plants in the Stillaguamish River Basin, Washington

## Introduction

Human activities in most areas of the developed world typically release nutrients, pharmaceuticals, personal care products, pesticides, and other contaminants into the environment, many of which reach freshwater ecosystems. In urbanized areas, wastewater treatment plants (WWTPs) are critical facilities for collecting and reducing the amounts of wastewater contaminants (WWCs) that ultimately discharge to rivers, coastal areas, and groundwater. Most WWTPs use multiple methods to remove contaminants from wastewater. These include physical methods to remove solid materials (primary treatment), biological and chemical methods to remove most organic matter (secondary treatment), advanced methods

to reduce the concentrations of various contaminants such as nitrogen, phosphorus and (or) synthetic organic compounds (tertiary treatment), and disinfection prior to discharge (Metcalf and Eddy, Inc., 1979). This study examined the extent to which 114 organic WWCs were removed by each of three WWTPs, prior to discharge to freshwater and marine ecosystems, in a rapidly developing area in northwestern Washington State. Removal percentages for each WWC were estimated by comparing the concentrations measured in the WWTP influents with those measured in the effluents. The investigation was carried out in the 700-mi<sup>2</sup> Stillaguamish River Basin, the fifth largest watershed that discharges to Puget Sound (fig. 1).



**Figure 1.** Locations of the wastewater treatment plants sampled in the Stillaguamish River Basin, 2009–11. (Modified from Wagner and others, 2014.)

**Table 1.** Characteristics of the wastewater treatment plants sampled in the Stillaguamish River Basin, 2009–11.

[Wastewater treatment plant: Years of sample collection are in parentheses. mm, millimeter]

Wastewater treatment plant characteristic	Wastewater treatment plant (year of sampling)			
	City of Arlington		Angel of the Winds Casino (2011)	Warm Beach Camp (2009)
	(2009)	(2011)		
Capacity				
Type and size of population served	Municipal; 17,711 residents	Municipal; 17,930 residents	Recreation; 3,600 visitors per day	Recreation; 800 seasonal visitors
WWTP capacity, in million gallons per day (number of connections)	1.00 (4,347)	2.65 (4,427)	0.18 (3)	0.035 (350)
Treatment processes				
Primary (solids removal)	6 mm screen; grit removal	3 mm screen; grit removal	2 mm screen; no grit removal	6 mm screen; no grit removal
Secondary (organic matter reduction)	Aeration basins	Aeration basins	Aeration basins	Two aerated lagoons followed by two constructed wetlands
Tertiary (additional contaminant removal)	None	Membrane bioreactor; biological nutrient removal	Membrane bioreactor	
Disinfection	Ultraviolet irradiation		Chlorination	Chlorination/ dechlorination
Point of discharge				
	Stillaguamish River		Groundwater recharge	Port Susan



City of Arlington WWTP (photograph taken by Ken Clarke, City of Arlington).



Angel of the Winds Casino WWTP (photograph montage assembled from Google Earth™ by Jack Barbash, July 2, 2014).



Warm Beach Camp WWTP (photograph from Dawda, 2013).

## Study Sites and Methods

The locations of the three WWTPs are shown in figure 1. The major water-treatment processes used by the three facilities are summarized in table 1. As indicated in table 1, prior to the 2011 sampling, the City of Arlington WWTP underwent a major upgrade that included the addition of a membrane bioreactor (MBR) and biological nutrient removal (BNR) to enhance contaminant removal. At each of the three WWTPs, paired samples of the influent and effluent were collected every 6 hours and composited over the same 24-hour period by the WWTP operators or by the Stillaguamish

Tribe of Indians. The composited influent and effluent samples were filtered and processed by the U.S. Geological Survey (USGS), and analyzed for their chemical composition at the USGS National Water-Quality Laboratory. The procedures used to characterize the physical and chemical conditions of the sampled waters, and to collect, preserve, and chemically analyze the samples, are described by Wagner and others (2014).

Removal percentages for each WWC were computed using the approach described by the U.S. Environmental Protection Agency (2010). For WWCs that were detected in the influent of a WWTP but not in the effluent, the removal percentages reported here represent minimum estimates. When this occurred for the two

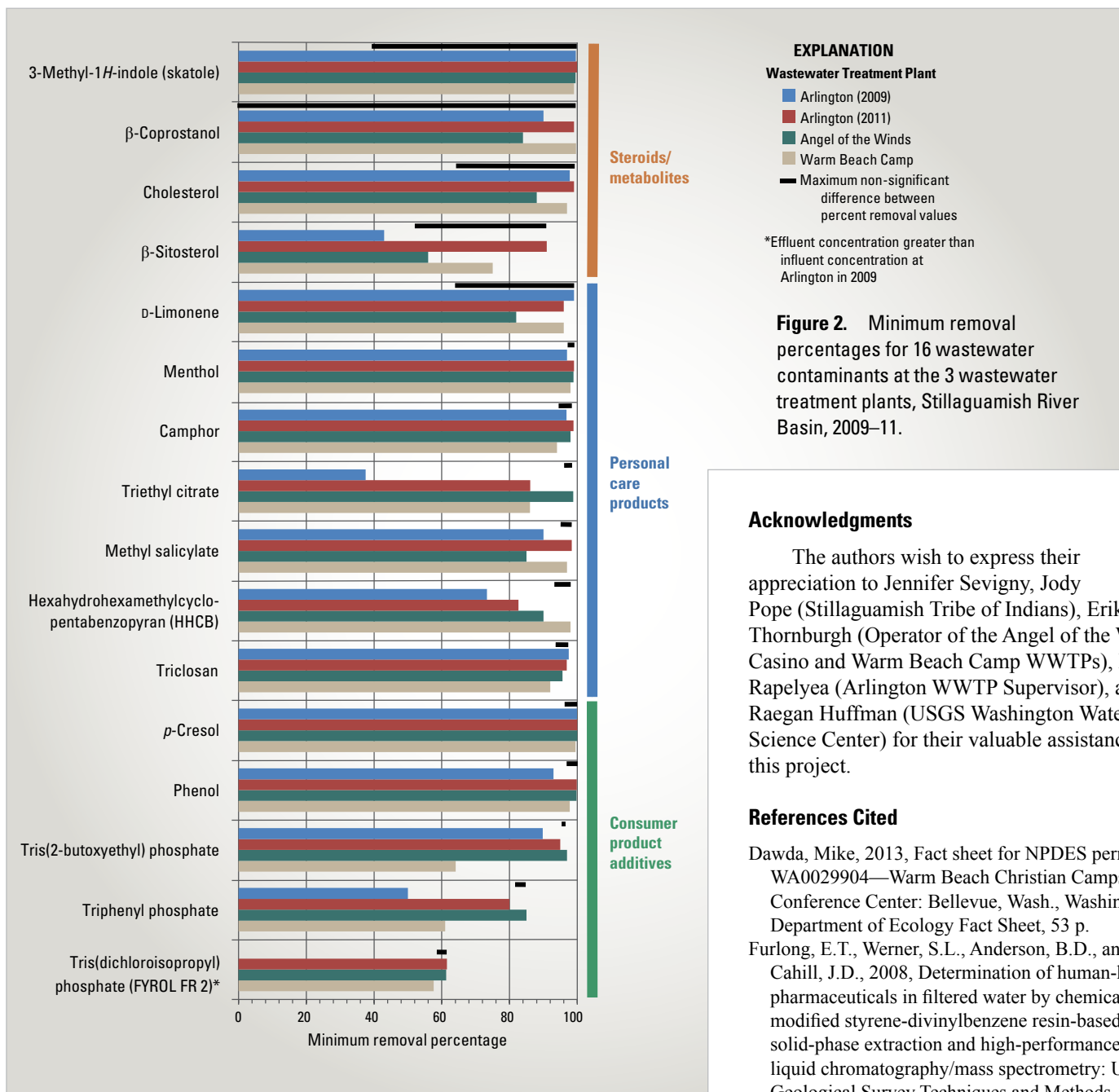
WWCs that were detected in field blanks at concentrations exceeding their reporting limits (benzophenone and camphor), the minimum removal percentages were computed using the maximum field blank concentration, rather than the reporting limit. The maximum non-significant difference (MNSD) between removal percentages for a given WWC ( $\alpha = 0.05$ ) was computed using the results reported from the analysis of spiked replicates of surface-water samples for the analytical method used for the compound of interest (Furlong and others, 2008; Zaugg and others, 2006, 2014).

## Contaminant Removal by the Wastewater Treatment Plants

Removal percentages are shown in figure 2 for the 16 WWCs for which removal percentages could be computed for all three WWTPs. The study results for the remaining 98 compounds are summarized in table 2, which shows the removal percentages for individual contaminants at the individual facilities. Table 2 also identifies instances where (1) the WWC was not detected in the influent; (2) the effluent concentration was greater than the influent concentration; (3) the chemical analyses were inconclusive because interfering substances were present, the compound was detected in the influent at its reporting limit, or the compound was not detected in the effluent using a reporting limit that was greater than the measured influent concentration; or (4) the analyses were incomplete because one of the samples (influent or effluent) was not analyzed, the maximum analyte recovery was less than 35 percent for one or more of the WWTPs, or at least one of the samples was not filtered prior to analysis.

Among the 114 WWCs examined for this study, 71 were detected in the influent of 1 or more of the 3 WWTPs. Of the 52 WWCs for which removal percentages could be computed at 1 or more of the WWTPs, nearly all the compounds





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(51) exhibited removal percentages greater than zero, and most compounds (49) showed removal percentages greater than 50 percent at 1 or more of the facilities. (Confidence in the removal percentages at the WWTPs examined for this study, however, was constrained because the influent and effluent for each facility were sampled only once in each year of interest.) The data shown in figure 2 and table 2 also indicate that after the MBR and BNR were added to the City of Arlington WWTP, significantly higher removal percentages were observed for eight compounds, and none showed a significantly lower removal percentage. (It was not possible to identify increases or decreases in removal percentages for a given compound after the MBR and BNR were installed if the removal percentages before and after the MBR and BNR installation were both minimum values—as was the case for  $\beta$ -sitosterol—or if only one removal percentage was a minimum value, but less than the other, as was the case for benzophenone.) The results from this study indicate that the water treatment processes used by these three WWTPs reduce the concentrations of many WWCs in the wastewater from the communities served by these facilities prior to discharge into the hydrologic system.

**Table 2.** Removal percentages for 92 wastewater contaminants for which removal percentages could not be computed for one or more of the wastewater treatment plants studied in the Stillaguamish River Basin, 2009–11.

[Shading—Tan, not detected in influent; green, effluent concentration greater than influent concentration. Not detected in influent or effluent of any WWTP: 5-Methyl-1*H*-benzotriazole; 9,10-Anthraquinone; Carbazole; 4-Cumylphenol; 4-*n*-Octylphenol; Dehydronifedipine. INC, chemical analysis inconclusive; MNSD, maximum difference between removal percentages considered non-significant ( $\alpha = 0.05$ ); WWC, wastewater contaminant; WWTP, wastewater treatment plant; >, greater than; –, chemical analyses incomplete or MNSD value not needed]

WWC	WWC removal percentage at WWTP					MNSD	WWC	WWC removal percentage at WWTP					MNSD
	City of Arlington		Angel of the Winds Casino (2011)	Warm Beach Camp (2009)				City of Arlington		Angel of the Winds Casino (2011)	Warm Beach Camp (2009)		
	(2009)	(2011)						(2009)	(2011)				
Consumer product additives							Pharmaceuticals—Continued						
1,4-Dichlorobenzene	60		—		—		Amitriptyline	—		—	—	—	
3-tert-Butyl-4-hydroxyanisole	—	—	—	INC	—		Antipyrine	—			—	—	
Acetophenone		> 50	> 7		—		Butalbital			—	—	—	
Acetyl hexamethyl tetrahydro naphthalene (AHTN)	67	78	—	> 90	3.7		Caffeine	—	99.8	—	—	—	
Benzophenone	> 78	91		> 92	2.9		Carbamazepine	—		—	—	—	
Isoborneol	> 67		—	> 64	—		Carisoprodol	—		—	—	—	
Isophorone	53		—		—		Celecoxib	—		—	—	—	
Isopropylbenzene			—		—		Chloroxylonol	—	99		—	—	
Isoquinoline			—		—		Chlorpheniramine			—	—	—	
N,N-diethyl-m-toluamide (DEET)	95	98		94	4.7		Citalopram	—	71	58.4	—	2	
Tetrachloroethene	—	—	—		—		Codeine				—	—	
Tributylphosphate		75	—	> 70	—		Dextromethorphan	—		—	—	—	
Tris(2-chloroethyl) phosphate (FYROL CEF)	0	25		63	0.77		Diazepam				—	—	
Consumer product metabolites							Dihydrocodeine	—		—	—	—	
1,7-Dimethylxanthine	> 99	> 99	—	> 99	—		Diltiazem	—				—	
2-Ethyl-2-phenylmalonamide	—			—	—		Diphenhydramine	—		—		—	
4-Nonylphenols (sum of all isomers)	INC	> 67		> 74	—		Efavirenz	—		—	—	—	
4-Nonylphenol diethoxylates (sum of all isomers)	75	93	—	90	1.9		Fluconazole	—			—	—	
4-tert-Octylphenol diethoxylate			—		—		Griseofulvin	—		—	—	—	
4-tert-Octylphenol monoethoxylate	INC			> 62	—		Hydrocodone			2.00	—	—	
4-tert-Octylphenol	80	> 81			3.8		Ibuprofen	—	INC	—	—	—	
Cotinine	—	> 85	—	—	—		Iminostilbene	—		> 72	—	—	
Natural biochemicals							Lidocaine	—			—	—	
β-Stigmastanol	INC	> 67	—	> 44	—		Meperidine	—		—	—	—	
Indole	—	> 99	—		—		Meprobamate	—	> 88		—	—	
Pesticides							Metaxalone			—	—	—	
Bromacil			—		—		Methadone			78	—	—	
Carbaryl			—		—		Methocarbamol	—	—	83.3	—	—	
Chlorpyrifos			—		—		Methylphenidate	—			—	—	
Diazinon	—		—		—		Oxycodone	67	79	77.9	—	1.9	
Metaxyl			—		—		Pentobarbital	—			—	—	
Metolachlor			—		—		Pentoxifylline	—		—	—	—	
Piperonyl butoxide	—	> 80	> 87	—	—		Phendimetrazine			—	—	—	
Prometon			—		—		Phenobarbital	—			—	—	
Petroleum-derived compounds							Phenytoin	—		—	—	—	
1-Methylnaphthalene	INC		—		—		Primidone	—	57	59	—	85	
2,6-Dimethylnaphthalene	INC		—		—		Propofol	—	76	—	—	—	
2-Methylnaphthalene	INC		—		—		Sulfamethoxazole					—	
Anthracene			—		—		Temazepam	—			—	—	
Benzo[a]pyrene	—		—		—		Thiabendazole	—		—	—	—	
Fluoranthene			—		—		Ticlopidine	—			—	—	
Naphthalene	INC	84	—		—		Tramadol	—	68		—	—	
Phenanthrene			—	INC	—		Trimethoprim		> 63	—	> 87	—	
Pyrene			—		—		Venlafaxine	—			—	—	
Pharmaceuticals							Verapamil	—			—	—	
Acetaminophen	—	> 99.9	—	—	—		Warfarin	—		—	INC	—	
Albuterol	—			—	—		Pharmaceutical metabolites						
							Chirald	—			—	—	
							Norpropoxyphene	—		—	—	—	
Trihalomethanes													
							Tribromomethane			—		—	

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