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Contaminants of emerging concern in a large temperate estuary☆

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ABSTRACT

This study was designed to assess the occurrence and concentrations of a broad range of contaminants of emerging concern (CECs) from three local estuaries within a large estuarine ecosystem. In addition to effluent from two wastewater treatment plants (WWTP), we sampled water and whole-body juvenile Chinook salmon (*Oncorhynchus tshawytscha*) and Pacific staghorn sculpin (*Leptocottus armatus*) in estuaries receiving effluent. We analyzed these matrices for 150 compounds, which included pharmaceuticals, personal care products (PPCPs), and several industrial compounds. Collectively, we detected 81 analytes in effluent, 25 analytes in estuary water, and 42 analytes in fish tissue. A number of compounds, including sertraline, triclosan, estrone, fluoxetine, metformin, and nonylphenol were detected in water and tissue at concentrations that may cause adverse effects in fish. Interestingly, 29 CEC analytes were detected in effluent and fish tissue, but not in estuarine waters, indicating a high potential for bioaccumulation for these compounds. Although concentrations of most detected analytes were present at relatively low concentrations, our analysis revealed that overall CEC inputs to each estuary amount to several kilograms of these compounds per day. This study is unique because we report on CEC concentrations in estuarine waters and whole-body fish, which are both uncommon in the literature. A noteworthy finding was the preferential bioaccumulation of CECs in free-ranging juvenile Chinook salmon relative to staghorn sculpin, a benthic species with relatively high site fidelity.

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1. Introduction

Contaminants of emerging concern (CECs) constitute a wide range of chemicals for which there is limited data on occurrence, environmental fate, and toxicity. Represented in this class of environmental contaminants are pharmaceutical and personal care products (PPCPs) and a number of industrial compounds such as polybrominated diphenyl ethers (PBDEs), perfluorinated compounds (PFCs), alkylphenols, bisphenol A, phthalates, and current-use pesticides. Many of these compounds are present in our rivers, estuaries, and coastal areas from wastewater treatment plant (WWTP) effluent discharging via outfalls to these water bodies. Other sources of CECs to waterways include discharges from industrial sources and aquaculture operations, in addition to runoff from impervious surfaces, landfills, biosolids application, and agricultural and farming activities (Gaw et al., 2014).

Most of these CECs are potent human and animal medicines that are used for various purposes, many of which are then excreted as the parent compound or as metabolites that flow into WWTPs. Some of these compounds are eliminated or reduced in concentration by treatment practices that vary among facilities or are sorbed to biosolids and removed from the waste stream (Lubliner et al., 2010; Oulton et al., 2010). By contrast, some CECs are poorly removed by WWTP processing or are discharged to surface waters, including streams, estuaries, or open marine waters due to secondary bypass or combined sewer overflows (Lubliner et al., 2010; Phillips et al., 2012).

There are several important factors to consider in assessing the environmental risk of CECs in estuarine waters, as well as other aquatic habitats. These include: the extent of product usage among local human populations, physical-chemical parameters (i.e. water

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solubility, hydrolysis, photodegradation, and adsorption to sediment and biosolids), rates of bioaccumulation, chemical potency, and potential toxicity to aquatic organisms and aquatic-dependent wildlife. Among these aforementioned factors, bioaccumulation and comparative toxicity to aquatic species constitutes the largest data gap in assessing ecological risk.

Over 4000 approved drug products are currently available (U.S. Food and Drug Administration, 2015) under various formulations and approximately 1100 are unique prescription and over-the-counter compounds comprising a large number of chemical classes and mechanisms of action (MoA). A consensus value of 324 drug targets has been proposed by Overington et al. (2006) for all classes of therapeutic drugs. A recent study of 12 fish species from a variety of families concluded that 65–86% of human drug targets are conserved in diverse fish species (Brown et al., 2014); therefore it is reasonable to assume that many of these drugs will also affect fish. Of the hundreds of chemicals that are likely present in the Puget Sound ecosystem, only a small percentage are currently monitored or regulated and there is little or no environmental toxicity information for the vast majority of these compounds. Many of these are common household chemicals that pass through wastewater treatment, have been approved for use and/or consumption by the general public, and are generally considered to be non-toxic. However, the higher-than-expected levels for some of these chemicals in aquatic organisms and possibly aquatic-dependent wildlife along with critical gaps in toxicological and risk assessment data underscores their importance for further investigation in the context of environmental and public health concerns (Roos et al., 2012; Arnold et al., 2014).

Relatively comprehensive analyses of CECs in the marine or estuarine ecosystem within the United States are uncommon. Notable exceptions for U.S. waters include the analysis of CECs in effluent and marine waters in southern California (Vidal-Dorsch et al., 2012) and Charleston Harbor (Hedgespeth et al., 2012), receiving waters in four estuaries along the Texas coast (Scott et al., 2015), San Francisco Bay (Klosterhaus et al., 2013), and Lubliner et al. (2010) who reported on effluent concentrations from WWTPs in Puget Sound, Washington. As far as we know, there are no studies that tested for a large suite of CECs in whole-body fish in marine waters.

Our approach in the present study involved a review of the literature that resulted in a prioritized list of 102 PPCPs, 17 hormones, and 31 industrial compounds to serve as a representative subset of CECs that we identified as a potential concern in the estuarine waters of Puget Sound, Washington, USA. Our primary goal was to determine the occurrence and concentrations of CECs in WWTP effluent, estuary water, and two fish species occupying different habitats with different life histories and compare among locations and matrices.

2. Methods

2.1. Selection of field sites

We selected three local estuaries as focal points for our study, including two estuaries that receive effluent from WWTPs and one as a reference site that is not known to have direct inputs from WWTP effluent. One contaminated site was Sinclair Inlet, which receives effluent from the Bremerton Westside WWTP (Fig. 1). The effluent outfall is located approximately 170 m from shore at a depth of 10 m below mean lower low water (MLLW) in in this local estuary. Sinclair Inlet has one other known source of effluent from the South Kitsap Water Reclamation Facility with a design flow of 16 million liters/d (MLD) (South Kitsap Water Reclamation Facility, 2013). The other contaminated site selected was the Puyallup River estuary, which receives effluent from the Tacoma Central WWTP (Fig. 1). The discharge outfall is at 40 m MLLW and approximately 370 m northwest from the mouth of the Blair Waterway in Commencement Bay. The Puyallup River basin contains 8 additional WWTPs with a combined permitted effluent volume of 63 MLD, with flows generally running much lower (Pierce County, 2010). The Nisqually estuary was selected as a minimally-contaminated reference site, and has been used in numerous studies as a reference site (as summarized by Meador, 2014). Table 1 contains additional details for each site.

Two fish species that commonly occur in Puget Sound estuaries were selected for assessing bioaccumulation of CECs. Specifically, Pacific staghorn sculpin (*Leptocottus armatus*) was selected for biomonitoring because this species is found widely in Puget Sound and U.S. west coast waters, generally exhibits high site fidelity, and may reside in estuaries for extended periods (Tasto, 1976). Juvenile Chinook salmon (*Oncorhynchus tshawytscha*) were selected based on their residence time (up to several weeks) in local estuaries where contaminants are often concentrated (Healey, 1991). Chinook salmon were selected over other salmonids that do not exhibit this life history trait (Meador, 2014). We also collected hatchery-reared juvenile Chinook salmon from the Voight's Creek hatchery on the Puyallup River for comparison to fish collected in the estuary. Fish were collected under a Washington State Scientific Collection Permit 13–046 and ESA Section 10(a) (1) (A) permit 17798. All methods for obtaining, transporting, and tissue sampling of fish were approved by the University of Washington Institutional Animal Care and Use Committee (protocol number 4096–01). Details of all sampling methods used in this study are reported in Yeh et al. (2013).

2.2. Sampling for CEC analytes in WWTP effluents and water

The effluent from Bremerton West WWTP was sampled on 9 September 2014 and the effluent flow was 13.2 MLD. The maximum monthly design flow from October–April is stated to be 58.7 MLD and permitted at 86 MLD (Bremerton Westside Factsheet, 2013). The effluent from Tacoma Central WWTP, Tacoma, WA was collected on 17 September 2014 and the flow on that day was 56.8 MLD. The maximum month design flow for wet weather is listed as 143.8 MLD (Tacoma Central WWTP Factsheet, 2004) and the permitted capacity is 228 MLD (Pierce County, 2010). These values do not include secondary treatment bypass during high volume flows or peak flows, which may exceed average flows by 2-fold. For the two week period prior to sampling, Tacoma experienced 0.03 inches of rain and Bremerton received 0.25 inches of rain (Weatherunderground, 2015).

At each WWTP, a total of 11 one-liter amber glass bottles were filled with effluent sampled at the final stage of processing, just before discharge into the outfall leading to the estuary. Similarly, at each field site a total of 11 one-liter amber glass bottles were filled with estuarine water at a depth of 2 m below the surface with a swing-sampling pole designed to collect water below the surface. We generally followed Washington Department of Ecology (2006) for obtaining water samples. Estuary water quality parameters including dissolved oxygen, conductivity, salinity, and temperature of the water column were measured at a depth of 2 m below the surface using the YSI Model 85 handheld probe (YSI Incorporated, Yellow Springs, OH). Similarly, the pH of the water column was measured using the Eutech Multi-Parameter PCSTestr 35 (Oakton Instruments, Vernon Hills, IL). One water sample was taken at each site and the estuary parameters were measured within minutes of water collection. No field blanks were collected.