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Spatial and temporal variability of contaminants within estuarine sediments and native Olympia oysters: A contrast between a developed and an undeveloped estuary

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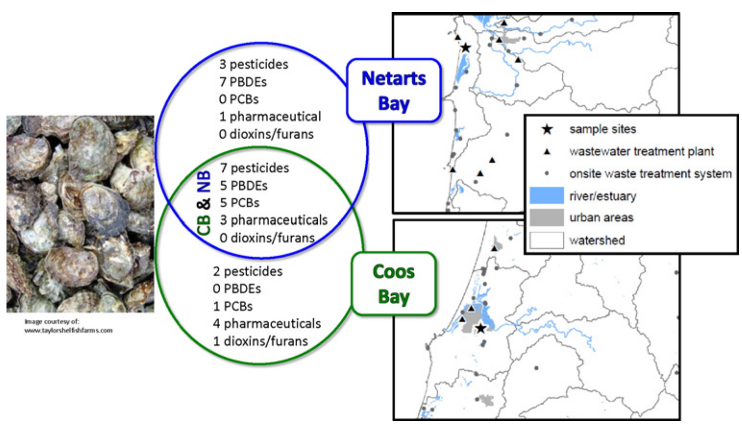
HIGHLIGHTS

- Spatio-temporal variability in contaminant deposition and uptake poorly known
- Seasonal and spatial sampling of oysters (biosphere) for contaminants (anthroposphere)
- Sediment (lithosphere) sampled to identify biota-sediment accumulation factors
- Emerging and legacy contaminants varied spatially and by season (anthroposphere).

GRAPHICAL ABSTRACT



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ABSTRACT

Chemical contaminants can be introduced into estuarine and marine ecosystems from a variety of sources including wastewater, agriculture and forestry practices, point and non-point discharges, runoff from industrial, municipal, and urban lands, accidental spills, and atmospheric deposition. The diversity of potential sources contributes to the likelihood of contaminated marine waters and sediments and increases the probability of uptake by marine organisms. Despite widespread recognition of direct and indirect pathways for contaminant deposition and organismal exposure in coastal systems, spatial and temporal variability in contaminant composition, deposition, and uptake patterns are still poorly known. We investigated these patterns for a suite of persistent legacy contaminants including polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) and chemicals of emerging concern including pharmaceuticals within two Oregon coastal estuaries (Coos and Netarts Bays). In the more urbanized Coos Bay, native Olympia oyster (*Ostrea lurida*) tissue had approximately twice the

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Metals
PCBs
Pesticides
Pharmaceuticals

number of PCB congeners at over seven times the total concentration, yet fewer PBDEs at one-tenth the concentration as compared to the more rural Netarts Bay. Different pharmaceutical suites were detected during each sampling season. Variability in contaminant types and concentrations across seasons and between species and media (organisms versus sediment) indicates the limitation of using indicator species and/or sampling annually to determine contaminant loads at a site or for specific species. The results indicate the prevalence of legacy contaminants and CECs in relatively undeveloped coastal environments highlighting the need to improve policy and management actions to reduce contaminant releases into estuarine and marine waters and to deal with legacy compounds that remain long after prohibition of use. Our results point to the need for better understanding of the ecological and human health risks of exposure to the diverse cocktail of pollutants and harmful compounds that will continue to leach from estuarine sediments over time.

1. Introduction

Estuarine and marine ecosystems provide the foundation for an invaluable suite of ecosystem services, including provisioning, regulating, supporting, and cultural benefits (MEA, Millenium Ecosystem Assessment, 2005). Over 50% of the global population currently resides within 80 km of the coast, and ocean ecosystems are now recognized to simultaneously be of great importance and under increased pressure from human populations worldwide (UNEP, 2006). Pollutants including persistent legacy contaminants and those of emerging concern present an ongoing threat to marine ecosystems, and they have the potential to damage and degrade a variety of ecosystem benefits as well as pose a risk to human health (e.g., Fleming et al., 2006; Kimbrough et al., 2008). Persistent legacy contaminants such as dichlorodiphenyltrichloroethane (DDT) and polychlorinated biphenyls (PCBs) remain in the environment long after they are introduced and several congeners are potentially carcinogenic to humans and animals. Contaminants of emerging concern (CECs) include pharmaceuticals, natural and synthetic hormones, and chemicals in personal care products that, historically, had less data available, but which, in recent years, have been recognized as potentially serious environmental threats, particularly to the endocrine and reproductive systems (Glassmeyer et al., 2005). For example, the sex ratio of white sucker fish (*Catostomus commersonii*) was biased toward females and a large proportion of intersex fish was observed downstream of wastewater treatment plant effluent that contained a mixture of hormones and hormone-mimicking surfactant metabolites (Vajda et al., 2008). Chronic exposure of a lake population of fathead minnow (*Pimephales promelas*) to low concentrations (5–6 ng/L) of the potent synthetic estrogen 17α-ethynylestradiol led to feminization of males, altered oogenesis in females, and ultimately, a near extinction of this species from the lake (Kidd et al., 2007).

Persistent contaminants and CECs may be introduced into the environment from a variety of sources. For example, although commercial use of PCBs is largely prohibited in the US, several uses are still authorized including incorporation into electrical and railroad transformers, natural gas pipelines, circuit breakers, and carbonless copy paper, and other applications (Oregon Department of Environmental Quality, ODEQ, 2003). Agricultural lands are an ongoing source of pesticides and herbicides, such as DDT and its breakdown products (dichlorodiphenyldichloroethane [DDD] and dichlorophenyldichloroethylene [DDE]), dieldrin, and chlordane. Household waste and dust are sources of polybrominated diphenyl ether (PBDE) flame retardants, which are used in motor vehicles, electronics and furniture (Schreder and La Guardia, 2014). Dioxins and furans are by-products emitted from a variety of industrial activities. Metals such as mercury may be transported to water bodies during wet deposition and overland runoff. Pharmaceuticals and personal care products (PPCPs) are discharged into the environment from households, hospitals, health-care facilities and other commercial operations. Consumers can excrete prescription or non-prescription pharmaceuticals at low concentrations, and excess pharmaceuticals are often flushed down the drain. Compounds in personal care products, such as alkylphenol-based surfactant metabolites present in soaps and detergents, are also washed away in sinks, showers, and lavatories. Consequently, wastewater treatment systems and onsite waste (i.e., septic) systems provide pathways of entry for these compounds into freshwater and marine systems (e.g., Paul et al., 1997; Conn et al., 2010). Several pharmaceuticals are also used to raise livestock; these compounds may be excreted by livestock and carried into rivers and streams (Barnes et al., 2002).

Little information is available on the occurrence of CECs and other contaminants in ecologically-sensitive areas such as estuarine and marine waters. Recent studies in Oregon revealed the presence of several different types of contaminants in the Willamette and Columbia Rivers which drain to the Oregon coast, within the Coos Estuary on the Oregon coast, and in wastewater effluent discharged into these waterbodies (Fig. 1; Hope et al., 2012; Morace, 2012; Nilsen et al., 2014; Pillsbury et al., 2015). However, very little information exists about contaminant concentrations in marine and estuarine organisms that occur in regions where the human population is low (i.e., less than 100,000 people along the entire Oregon coast). Historical sampling of marine organisms by the National Oceanographic and Atmospheric Administration (NOAA) Mussel Watch Program included mussel (*Mytilus* spp.) samples from six sites along the Oregon coast. This long term monitoring did not include CECs or Olympia oysters (*Ostrea lurida*; Kimbrough et al., 2008). Other coastal monitoring efforts such as EPA's National Coastal Condition Assessment do not include bivalves, which are important foundation species that filter feed and thereby accumulate chemical compounds. In the 1990s, sampling of softshell clams, oysters, and Dungeness crab in Coos Bay detected contaminants including butyltins and metals (ODEQ, personal communication).

Since exposure to multiple CECs with different modes of action may be a coast-wide ecological problem and potential human health issue, we examined the types and concentrations of contaminants and spatio-temporal variability of those compounds in two Oregon estuaries. Accumulation of persistent compounds and CECs in marine life has implications for ecological community structure, recreational and tribal shellfishing, and potential health consequences for human consumers (Guéguen et al., 2011; Gaw et al., 2014). The native Olympia oyster, the target species for this study, has drastically declined in abundance and distribution along the Pacific coast since the mid-1800s (White et al., 2009). In Oregon, several groups including NOAA, the South Slough National Estuarine Research Reserve, the Oregon Department of Fish and Wildlife, and The Nature Conservancy recently initiated actions to facilitate and guide restoration of Olympia oysters (Rumrill, 2010; Wasson et al., 2015). Recent sampling to examine CEC levels in Dungeness crabs (*Cancer magister*), California mussels (*Mytilus californianus*), and Oregon's coastal environment detected a suite of persistent legacy contaminants and CECs (Rodriguez del Rey, 2010; Granek, unpublished data).

Here we present new information about the chemical compounds and contaminant concentrations in sediment and oyster tissues from two Oregon estuaries that differ in the level of municipal, residential and industrial development. Our specific hypothesis is that persistent legacy contaminants and CECs will be elevated in sediments and oysters sampled from Coos Bay where municipal and residential development