Heavy metal and organic contaminants in oysters and sediments in resource waters, southeastern North Carolina

Project 2011-H-005

Submitted 6 December 2014

Final report to the Coastal and Recreational Fishing License (CRFL) Program
Marine Resources Fund
North Carolina Division of Marine Fisheries

Stephen A. Skrabal and Ralph N. Mead
Department of Chemistry and Biochemistry
University of North Carolina Wilmington

Abstract

Concentrations of various trace metals (most notably arsenic, copper, mercury, and zinc) and organic contaminants (polycyclic aromatic hydrocarbons and the antibacterial, triclosan) have been determined in oyster tissues and adjacent sediments in New Hanover and Brunswick counties, southeastern North Carolina. Trace metals that exceeded national median levels at multiple sites in this study included arsenic, copper, and zinc. Elevated levels of arsenic (exceeding the national median and, often, the national 85th percentiles) in oyster tissues are characteristic of much of the southeastern United States; these elevations are attributed to high natural background levels in the underlying bedrock and sediments as well as historical contamination by arsenic-containing agricultural pesticides. Less frequent copper elevations were mainly near marina activities, probably due to copper-containing antifouling paints. Zinc elevations were also localized, being most prominent near marinas and areas receiving roadway runoff. Another metal of national concern is mercury; however, concentrations of this metal were mostly at the national median for oyster tissue. There was generally little if any correlation between metal concentrations in oyster tissues and in adjacent surface sediments. Polycyclic aromatic hydrocarbons (PAHs) barely exceeded or were near the national median at only 3 sites, 2 in Lockwood Folly estuary, Brunswick County and 1 at Bradley Creek, New Hanover County. Concentrations at the remaining sites were 4 to >10 times less than the national median. The dominant source of the PAHs was combustion of fossil fuels. PAHs in oyster tissues tended to be dominated by lower molecular weight types. Triclosan, an antibacterial compound used in many consumer products, was found in oyster tissues and sediments at the 4 sites at which it was examined, including Bradley and Hewletts creeks which have experienced a number of sewage spills. Unexpectedly, it was also found at 1 site in Bald Head Island, a lightly populated area in Brunswick County. This indicates the widespread distribution of this common product. Oyster tissues contained triclosan at levels 2 to 43 times as high as adjacent sediments, indicating its bioaccumulation potential. Levels of metals and PAHs in oyster tissues are consistently elevated near more urbanized areas but are unlikely to be at levels harmful for human consumption.

INTRODUCTION

The eastern oyster (*Crassostrea virginica*) is an important fishery resource for North Carolina, accounting for annual average of $2.6 million in yearly sales between 2003 and 2013 in the state (NC Division of Marine Fisheries; *http://portal.ncdenr.org/web/mf/35-eastern-oyster-ssr-2013*). At the same time, oyster stocks have decreased precipitously over the past 125 years due to overfishing, declining water quality, and degradation and loss of habitat. In addition to their shellfishery value, oysters form reefs in intertidal and subtidal coastal waters. These reefs constitute ecologically valuable resources as they serve as habitat for a wide variety of fish and crustaceans, filter the water to improve clarity for submerged aquatic vegetation, and stabilize benthic regions against erosion. For these reasons, the combined economic and social impacts are much greater than simply the value of the harvest. Grabowski et al. (2012) estimate the value of oyster reefs in North Carolina at $5500 to $99,000 per hectare per year (2011 dollars), with much of the value resulting from shoreline stabilization. This can be compared to the gross value of the oyster harvest alone, which ranges from a few thousand to over $50,000 per hectare (2011 dollars) depending on the quality of the reef (Grabowski and Peterson, 2007; Grabowski et al., 2012). It is because of the various services of reefs that there have been concerted efforts over the past 20 years to restore degraded oyster reefs and to establish new ones to both serve as economic resources and to improve water quality

New Hanover and Brunswick counties (NHC, BC) in southeastern North Carolina have been active sites of oyster reef restoration, as the estuarine and tidal creek waters have widespread oyster reefs of varying quality. However, water quality in numerous locations throughout these two counties is negatively impacted as reflected in their inclusion on the 303(d) list (e.g., Lockwood Folly River (LFR), Northeast Cape Fear, portions of the Cape Fear River estuary) and/or in their closure to shellfishing, including 55% of shellfishing areas in Lockwood Folly (Lockwood Folly River Water Quality Strategy, 2007) and most tidal creeks in NHC. Systematic studies since 1993 have established that water quality in tidal creeks in NHC has been degraded by impacts related to increasing residential and commercial development (Mallin et al., 2008; also see reports at *www.uncw.edu/cmsr/aquaticecology/Laboratory/TidalCreeks/Index.htm)*. Water quality and habitat degradation is characterized by increased nutrient loadings, high fecal coliform inputs, occasional algal blooms, and sporadic hypoxia in the tidal creeks.

The relative amount of impervious surface in watersheds is a strong influence on the magnitude of degradation in tidal creeks and other water bodies in the southeastern US (Mallin et al., 2000; Holland et al., 2004). An increase in paved surfaces in watersheds allows increased runoff of municipal drainage water (Mallin et al., 1999, 2000, 2004; MacPherson et al., 2007). An episodic factor in water quality decline, particularly affecting NHC tidal creeks, has been raw sewage spillages caused by failed pipes and pumps (Mallin et al., 2007). Pulses of nutrients and fecal coliform from sewage spills appear to become attenuated relatively rapidly, probably due to biological uptake of nutrients by bordering marshes and settling of fecal coliform into sediments. A chronic problem in the LFR seems to be failing and poorly maintained septic systems which contribute fecal coliform and nitrogen-rich nutrients (Lockwood Folly River Water Quality Strategy, 2007). Water quality and habitat degradation in both NHC and BC have been clearly exacerbated by rapid population growth and increasing development in the past decade. Strong localized impacts have been noted for metal concentrations in the NHC tidal creeks, probably

due to inputs from marinas (especially for copper and zinc, used in antifouling bottom paints) and residential and commercial development (e.g., for arsenic and chromium used in treated lumber) (Burkhardt, 2009).

From 1993 to 2010, New Hanover County has ranked 2nd through 8th in emissions of a class of organic contaminants known as polycyclic organic matter, including polycyclic aromatic hydrocarbons (PAHs) and dioxins (*http://daq.state.nc.us/monitor/eminv/criteria*). PAHs are a class of organic compounds consisting of fused aromatic rings, produced by the incomplete combustion of biomass and fossil fuels. These compounds are highly hydrophobic and lipophilic, meaning that they strongly adsorb to suspended particles and sediments (as opposed to being dissolved in water) and tend to accumulate in fatty media including those in biological organisms. PAHs are most commonly introduced into the environment as stack effluents (as by the burning of coal or petroleum products) in the gas phase or adsorbed onto particulate matter and as runoff from paved surfaces containing the combustion byproducts of fuel and oil. PAHs are an ecological concern because of their toxicity to a wide variety of organism (including additive toxicity in PAH mixtures) and their tendency to bioaccumulate in tissues (Landrum et al., 2003).

Many impacted areas are also sites of water quality restoration efforts, including the LFR project headed by the North Carolina Coastal Federation and funded by the NC DWQ Section 319 program *(http://www.nccoast.org/Southeast/current-issues-lockwood.asp)*. The NCCF LFR project consisted of data collection for water quality parameters, a modeling component for fecal coliform, and a comprehensive watershed management implementation plan. The logical next step to successful implementation of the water quality improvement plan is an effort to reestablish oyster reefs and protect existing oyster resources, as outlined in several planning and management documents (NC DMF, 2001; Lockwood Folly River Water Quality Strategy, 2007; Oyster Restoration and Protection Plan for North Carolina, 2008). To this end, NCCF has recently been funded by the Estuary Restoration Act to begin oyster restoration efforts in LFR in spring and summer 2011. In NHC, oyster restoration efforts through the establishment and monitoring of oyster shell reefs have been implemented over the past 5 years in Bradley and Hewletts creeks, which flow into the Atlantic Intracoastal Waterway (AIW), and Dicks Bay, near Masonboro Island between the Atlantic Ocean and AIW (*http://www.nccoast.org/restoration-education/oyster-habitat.asp*).

Current efforts to monitor ecosystem health in the Cape Fear system using *C. virginica* have primarily relied on examining tissues for the presence of Dermo, an infection caused by the pathogenic protozoan, *Perkinsus marinus* (Couch et al., 2010). Heavy metals and organics, however, have been shown in numerous studies to be taken up by oysters with detrimental effects and to be present in the sediments comprising their habitat substrate (Wade et al., 1988; Presley et al., 1990; Guo et al., 2002, O'Connor, 2002, Kim et. al., 2008), although the exact relationship between oyster and sediment concentrations is not clear. Sediment studies on organics and metals in North Carolina estuaries suggest that effects on benthos may be localized (Hyland et al., 2000).

Comprehensive sampling of contaminants in oysters and sediments, however, has never been carried out in southeastern North Carolina at sufficiently high resolution to reliably evaluate their

impact on estuarine ecosystems in southeastern North Carolina, particularly with respect to current and planned restoration efforts. This lack of information is particularly striking given the known toxic effects that both excessive metals and organics can have on shellfish embryos, larvae, and adults (Martin et al., 1981; Sami et al., 1992). In addition, bivalves such as oysters have been widely recognized as effective sentinel species that reflect ambient water and sediment quality. Sentinel species tend to live and grow in one site for a significant period of time, inhabit areas that tend to be anthropogenically impacted, concentrate contaminants in their tissue by factors of 100-10,000 relative to water concentrations, only slowly excrete contaminants over time, and are valuable food and environmental resources (Farrington et al., 1983; O'Connor, 1998). Kim et al. (2008) examined relationships between a large suite of metals and organic contaminants and parasites and pathologies in bivalves, including oysters. They found that PAHs and heavy metals, particularly copper, zinc, silver, and lead, showed exhibited statistically significant relationships to various pathologic or parasitic conditions in oysters. This indicates the existence of complex, and as yet unknown, interactions between contaminants and oyster health.

Preliminary work at UNCW has documented levels of some metals and organic substances that may be at potentially harmful levels in tidal creek and estuarine environments in New Hanover County. In three tidal creeks (Bradley, Hewletts, Whiskey) creeks, sediments contained levels of Cu and Zn that exceeded the sediment LC_{50}, as determined in a previous study by Bat and Rafinelli (1998), for copper (Cu) at 7 of 11 sites and for zinc (Zn) at 8 of 11 sites for *Arenicola marina* (polychaete, "lugworm") and exceeded or were close to the limit for Cu and Zn at all sites for *Corophium volutator* (amphipod, "mud shrimp"). Preliminary studies for PAH distributions show increased levels in marsh sediments adjacent to roadways, suggestive of a runoff source.

OBJECTIVES

The goals of this project were to: (1) assess concentrations of selected trace metals and organic contaminants in sediments and oyster tissues in Lockwood Folly (BC), an impaired system undergoing restoration, and in tidal creeks and other potential oyster restoration areas in NHC; (2) partner with researchers with the NC Division of Marine Fisheries, UNCW, and the NC Coastal Federation in choosing sampling sites in NHC and Lockwood Folly that are currently undergoing oyster restoration or that will be restored in the future; (3) use oyster growth data and other indicators collected by these partners along with our oyster tissue metal and organic data to assess ecosystem and habitat health and restoration success; (4) work with UNCW undergraduates to present chemical data obtained in this study and that obtained in previous studies in an interactive Web-based application that will enable citizens, students, and other interested parties to view chemical data (with interpretation) for all sampled sites; and (5) provide scientific training at UNCW to undergraduate students (and potentially, high school students through the UNCW Marine Quest program) to improve their knowledge of methods of assessing ecosystem health using geochemical indicators.

The emerging contaminant triclosan (5-chloro-2-(2,4-dichlorophenoxy)phenol) is particularly interesting because it is an antibacterial and antifungal agent commonly added to a variety of

consumer products, including soaps, toothpastes, deodorants, and cleaning solutions. It has a strong tendency to accumulate in sediments and on particles and may persist in the environment for relatively long periods of time (Chau et al., 2008). Given the sources, it may be a particularly accurate indicator of human waste inputs, as from leaking septic systems, wastewater treatment plants, and other sewage sources. Triclosan and its breakdown products may pose sublethal effects on aquatic organisms at low concentrations (Sumpter and Johnson, 2005; DeLorenzo et al., 2008).

METHODS

Sampling

Each of 19 sites were sampled between September 2011 and September 2012 and were chosen so as to obtain a representative geographical representation of intertidal oyster reefs in NHC (15 sites) and LFR (4 sites) (Figure 1). In particular, NHC tidal creek sites normally included stations at the head and mouth of each system, and where applicable, proximity to current reef restoration projects. One Atlantic Intracoastal Waterway (AIW) site near the UNCW Center for Marine Science was also chosen because a number of field studies are ongoing at that location. Sites at the relatively small LFR were chosen to encompass the major reef systems; as stated previously, oyster reef restoration is ongoing in this system.

At each site, oysters (generally at least 2.5 cm in shell height) were collected by carefully separating individuals from reefs. Approximately ten individuals were collected for metals analyses and twenty for organic analyses at each site. Oysters were gently scrubbed to remove mud and placed in a bucket of ambient water and transported to the lab.

Surface sediment samples (top ~2 cm) were collected using a plastic spoon and placed in heat sealable Kapak bags. Three bags of sediment were collected from each site—one for sediment analysis, one for organic analysis, and one archive.

Sample processing

Bags containing the sediment samples were heat sealed and stored frozen until further processing. Oysters were allowed to depurate in ambient water for 24-48 hours. After depuration, oysters were scrubbed again and rinsed with deionized water, then stored frozen in plastic ziplock bags. Once thawed, the oysters were shucked using a stainless steel shucking knife. If excess particulates or other debris was found, high-purity deionized water (Milli-Q, >18.2 MΩ cm^{-1}, Waters Corp.) was used to rinse the tissue of contaminants. For metals analyses, the whole animal was carefully removed from the shells using a stainless steel scalpel and placed into individual pre-weighed, acid-washed 20-mL polyethylene vials. The samples were then frozen and freeze-dried using a Labconco Freezone 4.5 at -50° C and 0.021 mBar. The dry oyster tissue mass, as well as the mass and length of the dried shells, were determined. Dried samples were pulverized using a Teflon policeman inside the vial to reduce contamination as well as sample loss. For organic analyses, the meats from 20 oysters at each site were combined into a heat-sealable Kapak bag prior to freezing and freeze-drying. Samples for organics were then stored

sealed in these bags until further processing. Sediment samples for metals and organic analyses were freeze-dried, homogenized, gently disaggregated, and passed through a polyethylene/nylon sieve to obtain the <500 µm size fraction. The weight percent coarse (>500 µm) and fine (<500 µm) material were recorded for the metals samples.

Metal analyses

Oyster tissues were digested using standard techniques with mixed acids (USEPA 1992). Briefly, portions of typically 0.2 g were weighed into Teflon microwave digestion vessels (HP-500 vessels, CEM Corp.) and 5.00 mL of a 4:1 mixture of concentrated nitric and hydrochloric acids (OmniTrace, EMD) was added. The vessels were sealed and placed into a CEM MARS 5 microwave where digestion was carried out by ramping temperature to 180°C over 4 min and holding at this temperature for 11 min. At least one blank (consisting of the acid mixture only) and typically one sample of standard reference material (NIST 1566b oyster tissue) were run with each batch to assure quality. After solutions had cooled, they were quantitatively transferred to acid-washed 30-mL polyethylene bottles. Solution masses were recorded; these were converted to volumes by measuring the solution densities, which averaged 1.05 g mL^{-1}. Duplicate 1-g portions of each sediment sample were leached in 20 mL of 1 M hydrochloric acid (trace metal grade) for 2-h with continuous agitation (250 rpm on shaker table). Following treatment, samples were filtered through 0.45 µm cellulose acetate filters and stored in polyethylene bottles.

Oyster and sediment samples were analyzed for a suite of trace metals (arsenic, cadmium, cobalt, copper, lead, manganese, nickel, silver, and zinc using inductively-coupled plasma optical emission spectrometry (ICP-OES) and multi-element calibration curves prepared by diluting commercial high-purity 1000 ppm standards. Concentrations of mercury were determined on dry oyster tissues using a Milestone DMA-80 analyzer calibrated using dilutions of a certified 1000 ppm commercial standard. Metal concentrations in oyster tissues are only reported for metals which were recovered in acceptable levels (approximately 80-100%) in repeated analyses of the certified standard reference material NIST 1566b. Analytes that were consistently within the acceptable levels were arsenic, cadmium, cobalt, lead, manganese, mercury, nickel, and zinc. Silver was acceptable in only a selected subset of stations. Of these analytes, a subset (cadmium, cobalt, lead, nickel) showed concentrations that were only slightly above detectable levels and will be only briefly discussed. The remaining metals showed significant patterns that are discussed in more detail.

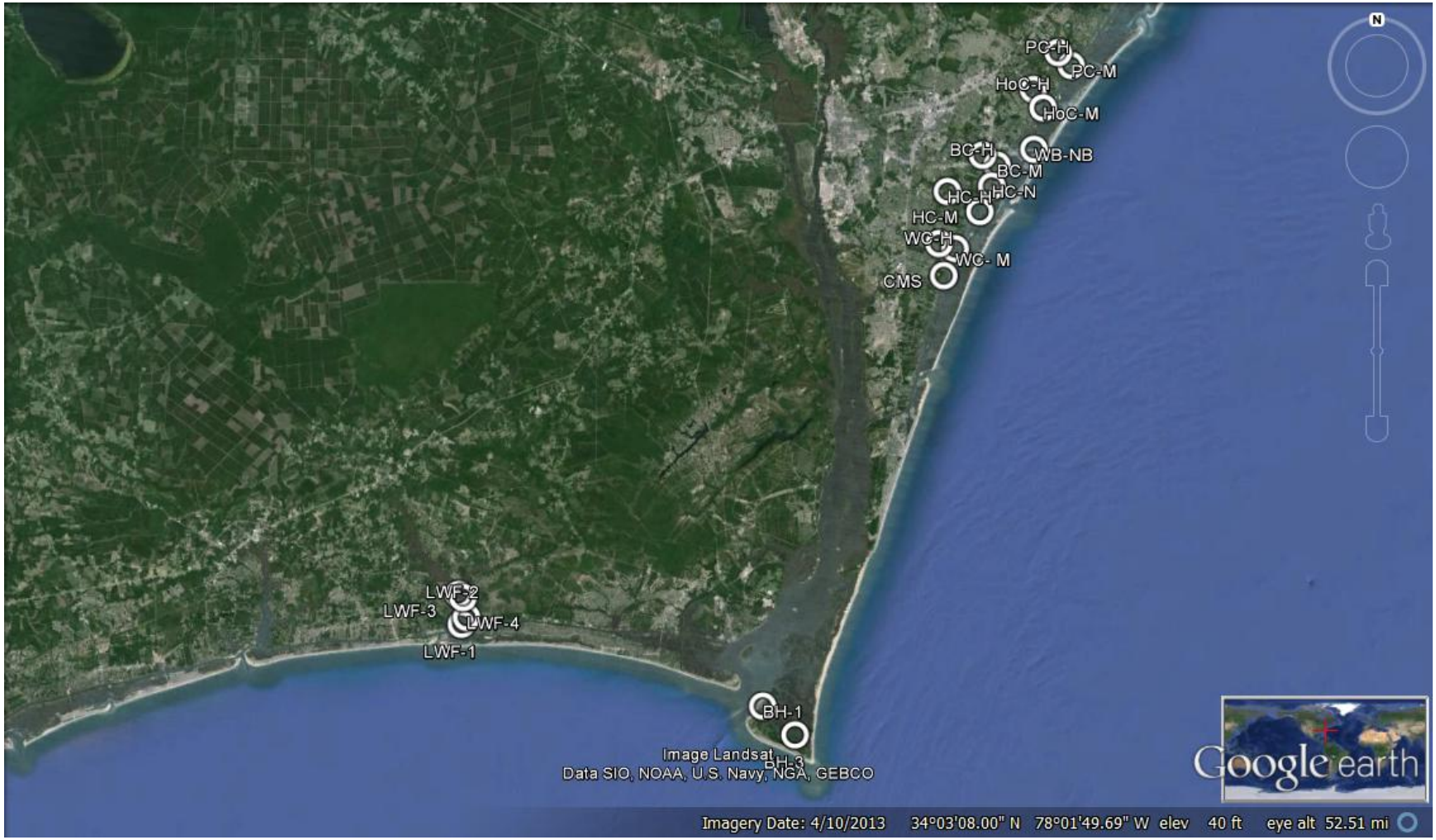


Figure 1. Sampling sites for oysters and sediments in New Hanover and Brunswick counties, North Carolina. Base image from Google Earth.

Organic analyses

Duplicate sediments and oyster tissues were analyzed for a suite of PAHs following EPA method 8100 (US EPA, 1986). Triclosan was analyzed using the method of Agüera et al. (2003). Briefly, dried and ground sample material was weighed and extracted in 100% methylene chloride in a Soxhlet extractor for 24 hours. The extract was then fractionated by silica gel chromatography into four fractions that contained PAHs and triclosan. These fractions were analyzed by gas chromatography/mass spectrometry and quantified using external calibration curves. Standard reference material (SRM) 1941b from National Institute of Standards and Technology (NIST) was used to validate the sediment PAH extractions whereas SRM 2974a was used to validate PAHs in mussels. These two standards were chosen to best mimic the matrix analyzed in this study. Each SRM was analyzed three times by the method above and the concentrations of PAHs were compared to the certified values. Overall, the sediment extraction and quantification provided between 80-100% of the certified values whereas the mussel PAHs were between 70-85% of the certified values. The reported organic concentrations were not corrected for recoveries and can be considered minimum values.

RESULTS

All sample information, including site names and locations, sample characteristics, and analytical data, are shown as an appendix (Table A1).

Oyster physical characteristics

Mean shell heights of oysters collected at each site ranged from 5.9 to 9.4 cm with considerable variability at each site (Figure 2a). Mean dry tissues weights varied between 0.15 and 2.3 g, with most weights falling between about 0.3 and 0.6 g (Table A1). At selected sites, the condition index (CI) of oysters was calculated using the formula suggested by Lawrence and Scott (1982):

$$CI = 100\ [\text{Dry tissue weight (g)} / (\text{Whole oyster weight (g)} - \text{shell weight (g)})].$$

The condition index has been used in a number of studies as a semi-quantitative means of assessing how much of the shell volume is occupied by tissue; consequently, a higher CI suggests a healthier oyster (Lawrence and Scott, 1982). This value can be affected by processing factors such as unremoved fouling on oyster shells, loss of shell material during shucking, incomplete removal of tissues from the valves, and by physiological factors such as environmental stress, and tissue loss during spawning in summer months (Riedel et al., 1998; Abbe et al., 2000).

Average values of the CI in this study ranged between 2.0 and 6.9 at all sites, with lower values at the Lockwood Folly sites (2.0-3.8) relative to all other sites (4.9-6.9) (Figure 2b). All sites were sampled in either March, April, or September, with the exception of sites HC-M, HC-N, PC-H, PC-M, and HoC-M, which were sampled in July. Apart from the LWF sites, the CI values are at the low end of the range of values reported over different seasons in the Patuxent estuary, Maryland (3-14, Abbe et al., 2000; 6.5-9, Riedel et al., 1998). The particularly low values at

LWF likely reflect loss of tissue during the spawning season, although this would have to be verified by seasonal studies.

Values for shell height and CI are informational for our sampled oysters and not necessarily meant to fully characterize oysters at site, as our sample design was not randomized nor carried out over different seasons.

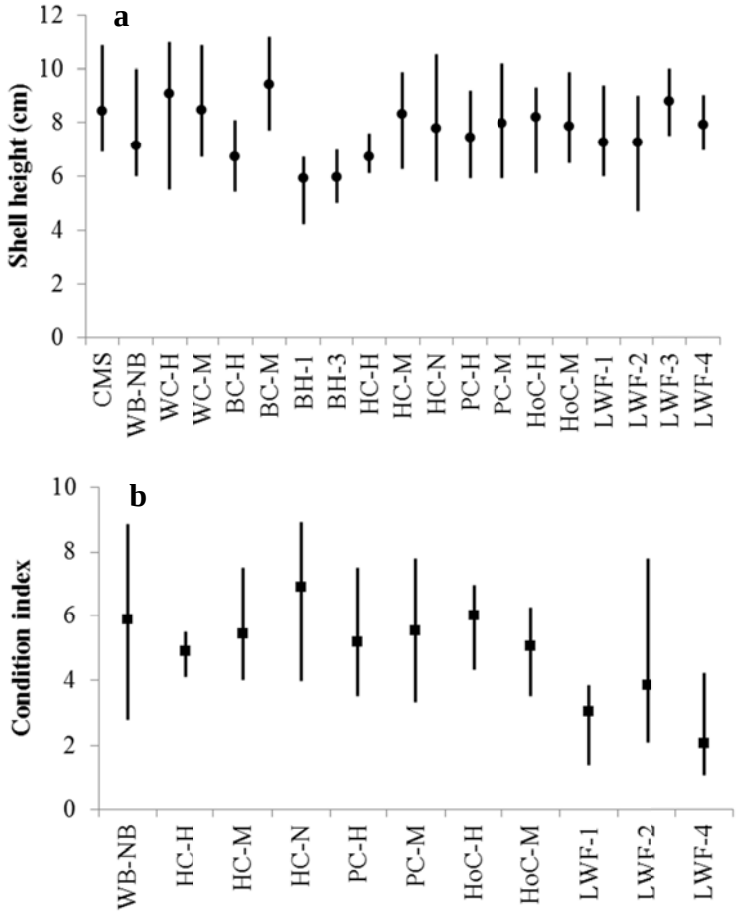


Figure 2. Means (symbols) and ranges (vertical lines) of (a) shell height and (b) condition index of oysters at each study site.

Metals in oyster tissues

Throughout this discussion, metal concentrations in oysters at each site will be discussed in terms of variations in individual oysters at each site and, in order to more validly compare concentrations between sites, as composite concentrations. The composite concentration (C_{comp}) estimates the concentration obtained if the individuals had been pooled into one sample, and is calculated as

$$C_{comp} = \Sigma (c_i m_i / \Sigma m_i)$$

where c_i is the concentration of a metal in an individual oyster and m_i is the sample mass.

Arsenic

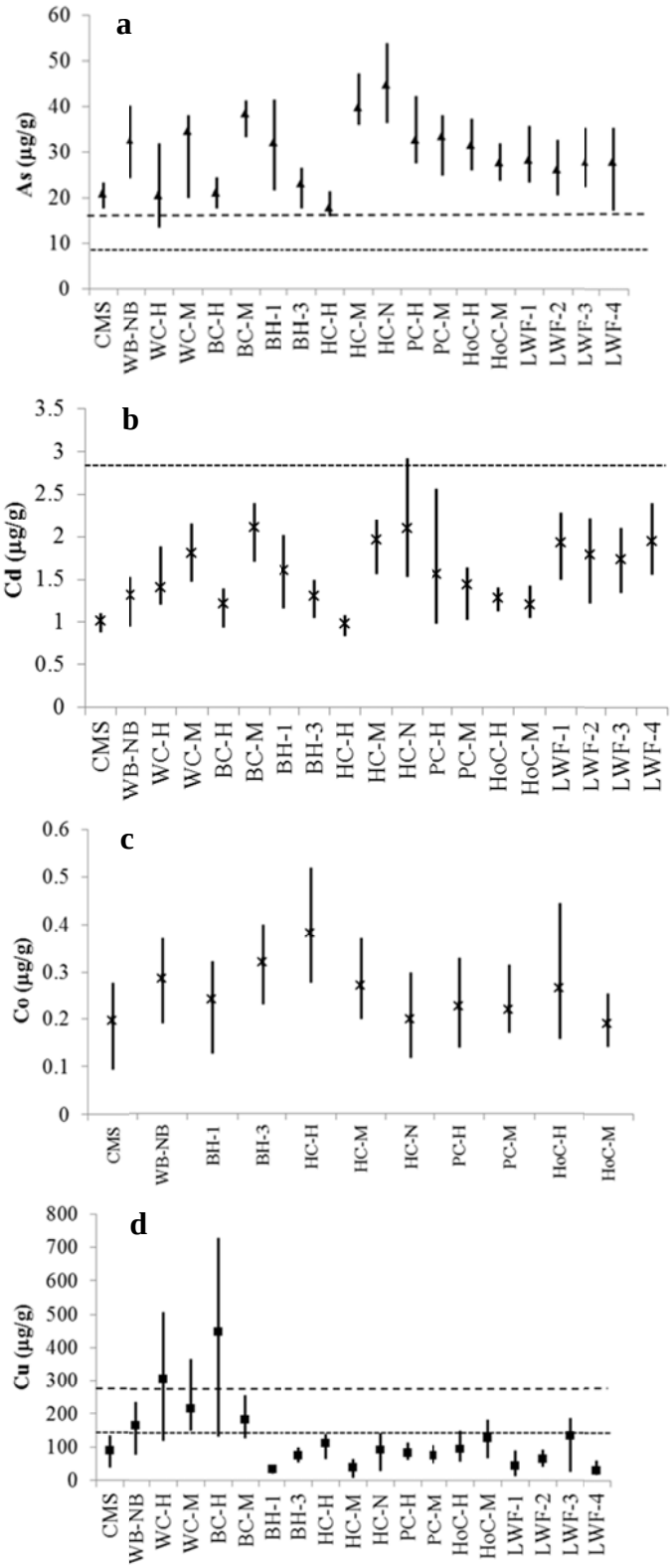
Oysters at all sites were analyzed for As; concentrations in individual oysters and composites ranged between 13-54 and 18-45 μg g^{-1}, respectively (Figure 3a). The highest composite concentrations (39-45 μg g^{-1}) were found at or near the mouths of Bradley and Hewletts creeks (sites BC-M, HC-M, HC-N). At the five tidal creeks where both head and mouth samples were obtained, As concentrations at the mouths were 1.5-2 times higher than at the heads of Whiskey, Bradley, and Hewletts creeks, whereas there was little difference between locations at Pages and Howe creeks. All oysters had As concentrations well above the national median (9.2 μg g^{-1}) and 85th percentiles (16 μg g^{-1}) (Lauenstein et al., 2002). Valette-Silver et al. (1999) report a median As value for the southeastern US of 20.6 μg g^{-1} based on the NOAA National Status and Trends (NS&T) program. Composite values at 17 of 19 sites in this study exceeded that value.

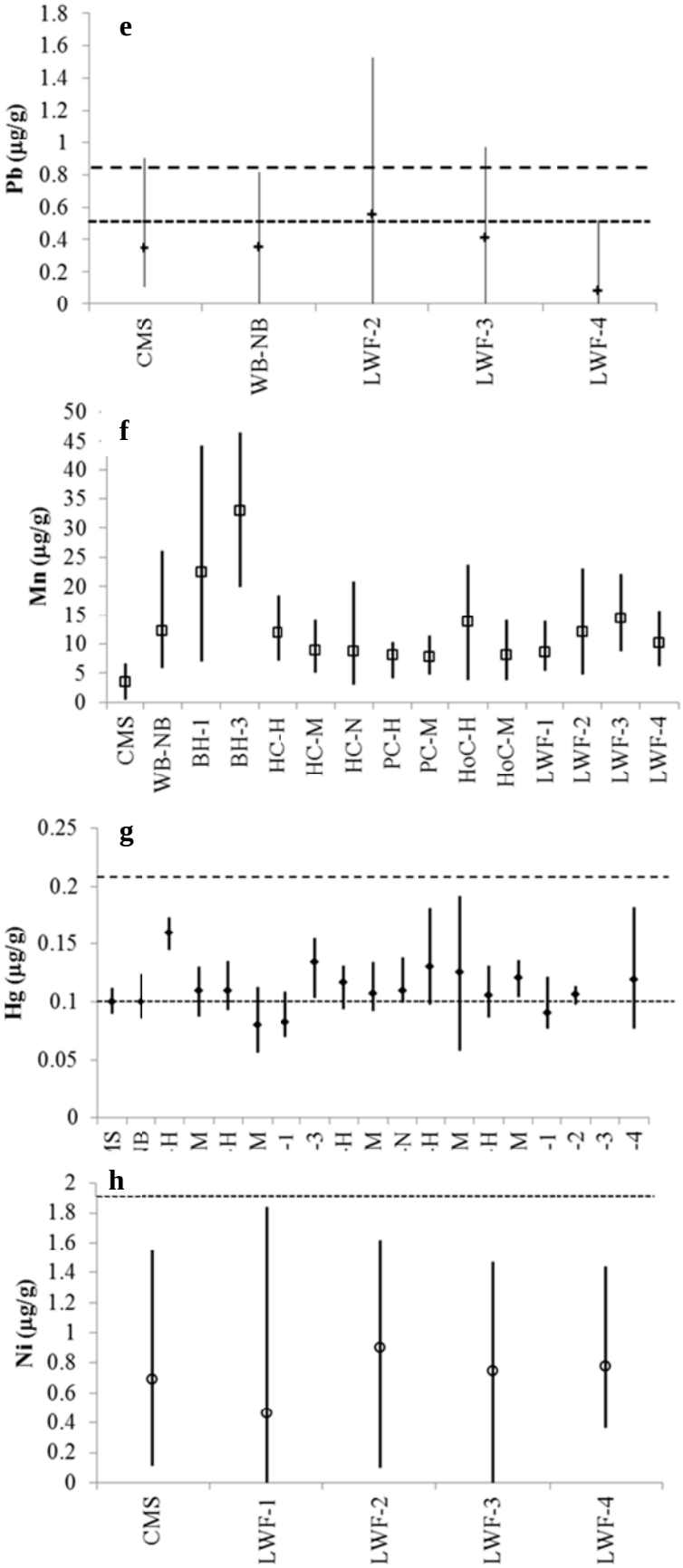
Copper

Concentrations of Cu at all 19 sites ranged widely from 7.4-729 μg g^{-1} in individual oysters and 29-444 μg g^{-1} in composites (Figure 3d). Concentrations were markedly higher near marinas and where numerous vessels were secured, particularly Bradley and Hewletts creeks. Concentrations of Cu in oysters were significantly larger at the heads of Whiskey, Bradley, and Hewletts creeks relative to the mouths (*t*-test of means, $p < 0.05$). This is tentatively attributed to the accumulation of fine sediments containing Cu in the heads of these creeks. Composite samples at three sites (WB-NB, WC-M, BC-M) exceeded the national median concentration of (140 μg g^{-1}) and two sites (WC-H, BC-H) exceeded the 85th percentile value of 290 μg g^{-1} (Lauenstein et al., 2002).

Lead

The overall low concentrations of Pb could only be confidently determined at 5 sites; these concentrations ranged from about 0.1 to 1.5 μg g^{-1} in individual oysters, with most concentrations, including the composites, <1 μg g^{-1} (Figure 3e). These limited values are similar to the national median for Pb (0.51 μg g^{-1}; Lauenstein et al., 2002) and similar to the range (0.2-1.5 μg g^{-1}) found in Galveston Bay (Jiann and Presley, 1997).


a
As (µg/g)
0
10
20
30
40
50
60
CMS
WB-NB
WC-H
WC-M
BC-H
BC-M
BH-1
BH-3
HC-H
HC-M
HC-N
PC-H
PC-M
HoC-H
HoC-M
LWF-1
LWF-2
LWF-3
LWF-4
b
Cd (µg/g)
0
0.5
1
1.5
2
2.5
3
3.5
CMS
WB-NB
WC-H
WC-M
BC-H
BC-M
BH-1
BH-3
HC-H
HC-M
HC-N
PC-H
PC-M
HoC-H
HoC-M
LWF-1
LWF-2
LWF-3
LWF-4
c
Co (µg/g)
0
0.1
0.2
0.3
0.4
0.5
0.6
CMS
WB-NB
BH-1
BH-3
HC-H
HC-M
HC-N
PC-H
PC-M
HoC-H
HoC-M
d
Cu (µg/g)
0
100
200
300
400
500
600
700
800
CMS
WB-NB
WC-H
WC-M
BC-H
BC-M
BH-1
BH-3
HC-H
HC-M
HC-N
PC-H
PC-M
HoC-H
HoC-M
LWF-1
LWF-2
LWF-3
LWF-4


e
Pb (µg/g)
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
CMS
WB-NB
LWF-2
LWF-3
LWF-4
f
Mn (µg/g)
50
45
40
35
30
25
20
15
10
5
0
CMS
WB-NB
BH-1
BH-3
HC-H
HC-M
HC-N
PC-H
PC-M
HoC-H
HoC-M
LWF-1
LWF-2
LWF-3
LWF-4
g
Hg (µg/g)
0.25
0.2
0.15
0.1
0.05
0
h
Ni (µg/g)
2
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
CMS
LWF-1
LWF-2
LWF-3
LWF-4

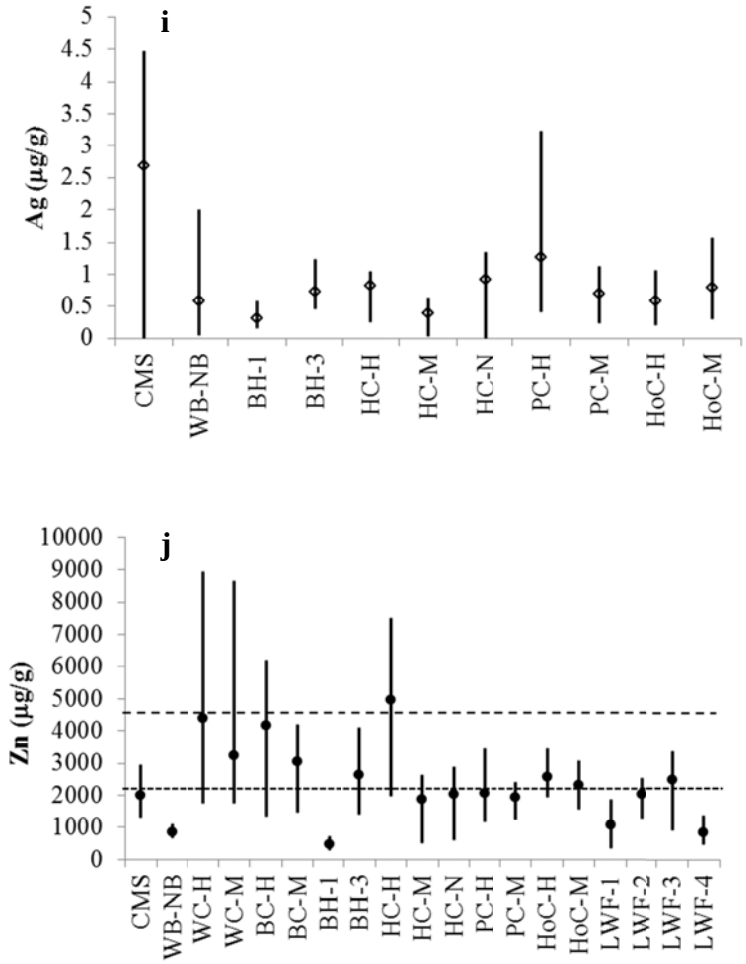


Figure 3. Composite concentrations (symbols) and range of individual oyster concentrations (solid vertical lines) at each sampling site. Dotted and dashed lines indicate national median and 85th percentile concentrations, respectively, for oysters reported in Lauenstein et al. (2002). Absence of lines indicate no national medians are reported for that metal. (a) arsenic, (b) cadmium, (c) cobalt, (d) copper, (e) lead, (f) manganese, (g) mercury, (h) nickel, (i) silver, (j) zinc.

Manganese

Manganese in generally not regarded as having significant pollutant sources except in specific settings. Concentrations of Mn in oyster tissues are likely to be dominated by naturally occurring suspended mineral matter rather than contaminant sources. Concentrations of Mn at 15 sites ranged between 0.5 and 47 μg g^{-1} in individual oysters and composites (Figure 3f). Most composite concentrations ranged between about 7 and 14 μg g^{-1}.

Mercury

Concentrations of Hg were determined at 18 sites; individual and composite concentrations ranged between 0.06 and 0.18 μg g^{-1} (Figure 3g). Composite concentrations were all centered around the national median concentration of 0.1 μg g^{-1} and no individual concentrations exceeded the 85th percentile level of 0.21 μg g^{-1} (Lauenstein et al., 2002). The form of Hg that is bioaccumulated in organisms, including a variety of fish species in North Carolina (Sackett et al., 2009) and worldwide is methylmercury, formed mainly in sedimentary environments by the actions of naturally-occurring sulfate-reducing bacteria. The total concentrations of Hg determined in these oyster tissues is assumed to be mainly in the form of methylmercury.

Nickel

Nickel could only be confidently determined at 5 sites (Figure 3h). Individual and composite concentrations ranged between 0.1 to 1.8 μg g^{-1}, similar to or lower than the national median (1.9 μg g^{-1}; Lauenstein et al., 2002).

Silver

Silver was determined at 11 sites (Figure 3i). Individual concentrations ranged between 0 (below detection, BD) and 4.5 μg g^{-1}, with the large majority of individual and composite values <2 μg g^{-1}. The exception was the CMS site, with individual concentrations of up to 4.5 μg g^{-1} and a composite of 2.7 μg g^{-1}. There is no national median for Ag; however, studies of Ag distributions in oyster tissues from relatively impacted estuaries show much higher concentrations (5 to 55 μg g^{-1} in Potomac estuary, Maryland (Daskalakis, 1996); <1-28 μg g^{-1} in Galveston Bay, Texas (Jiann and Presley, 1997); 1-11 μg g^{-1} in the Patuxent estuary, Maryland (Riedel et al., 1998)).

Zinc

Concentrations of Zn were determined at all 19 sites (Figure 3j). Individual and composite levels ranged between 291-8940 and 469-4950 μg g^{-1}, respectively. Composite concentrations at 8 sites exceeded the national median level of 2200 μg g^{-1} and 1 site exceeded the 85th percentile value of 4600 μg g^{-1} (Lauenstein et al., 2002). Levels of Zn were higher in the heads relative to the mouths of all tidal creek systems studied (Bradley, Whiskey, Hewletts, Pages, Howes creeks; *t*-test of means, $p < 0.05$). The highest concentrations of Zn (4180-4950 μg g^{-1}) were found at the heads of the mostly heavily developed creeks (Bradley, Hewletts, Whiskey creeks). This is consistent with the multiple sources of Zn including marinas and runoff from roads (Comber et al., 2002; Göbel et al., 2007). Interestingly, the Bald Head Island site BH-3, located in an interior

salt marsh, had a Zn composite concentration above the national median at 2650 μg g^{-1}. This elevation co-occurs with somewhat elevated PAH concentrations, as discussed later.

Metal concentrations vs. oyster physical parameters

The metals of greatest significance in the study are As, Cu, and Zn, because of multiple exceedances of the national median and/or the 85th percentile, and Hg, because of concerns regarding bioaccumulation of this particularly toxic metal. Correlations were examined between individual oyster tissue As, Cu, Hg, and Zn concentrations and shell height, which is an indirect measure of oyster age (Figure 4); and condition index, a measure of how much of the volume of the shell is occupied by tissue (Figure 5).

The significance of correlations was examined using a *t*-test of the Pearson product-moment correlation coefficients (*R*). A marginally significant positive correlation was found between As and shell height ($p = 0.0576$), whereas Cu, Hg, and Zn showed no significant correlation ($p = 0.108$, 0.334, 0.204, respectively). Significant positive correlations were found between CI and As ($p < 0.0001$), Cu ($p = 0.00333$), and Hg ($p = 0.0229$) whereas no significance was found for Zn ($p = 0.271$). These results suggest that older oysters (as parameterized by shell height) accumulate more As, whereas physiologically more robust oysters (as reflected in the CI) accumulate more As, Cu, and Hg.

Metal concentrations in oyster tissues vs. sediments

Correlations between the metal contents of oyster tissues relative to proximal sediments were analyzed using *t*-tests of the correlation coefficient (*R*), as described previously (Figure 6.). Only selected metals (As, Cd, Cu, Zn) for which adequate analyses were available were examined. A positive relationship is seen for Cu (Figure 6c, $p = 0.00709$), a weak inverse relationship is seen for Cd ($p = 0.0680$), and no relationship is observed for As and Zn ($p = 0.155$ and 0.289, respectively). No Hg analyses were available for sediments in this study; however, numerous analyses of the Hg content of sediments in New Hanover County have shown the concentrations to range predominantly between 0.01 and 0.4 μg g^{-1} (Skrabal, unpublished data) whereas composite oyster Hg concentrations range relatively narrowly between 0.08-0.16 μg g^{-1}.

Of the limited sediment metal analyses, the only sediment quality criteria exceedances were for As and Cu at site BC-H. Copper (36.9 μg g^{-1}) and As (8.07 μg g^{-1}) exceeded the threshold effects level (TEL; 18.7 μg g^{-1} for Cu, 7.24 μg g^{-1} for As), the concentration below which few adverse effects would be expected. These levels were well below the probable effects levels (41.6 μg g^{-1} for As, 108 μg g^{-1} for Cu (MacDonald et al., 1996), where the PEL is the concentration above which adverse effects would be frequently observed.

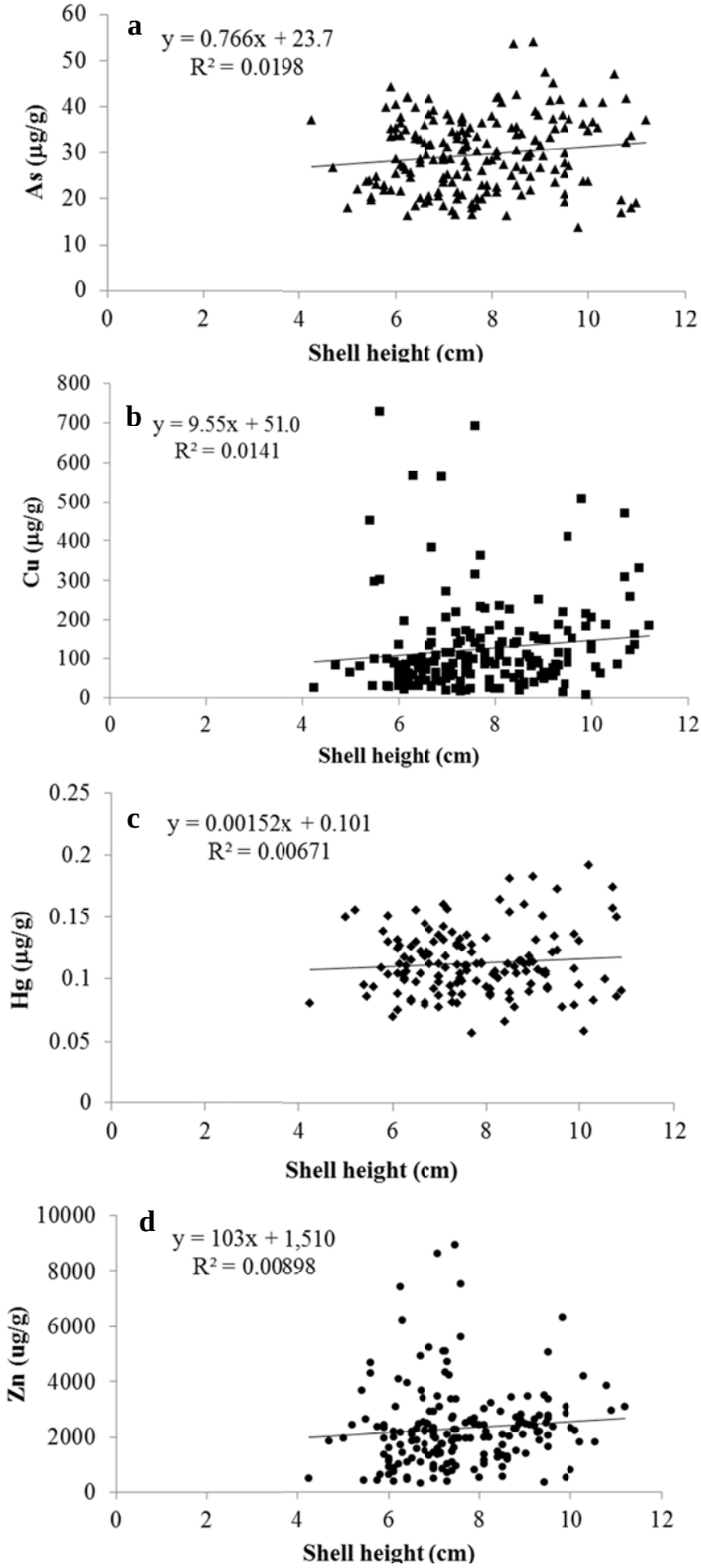


Figure 4. Correlations between shell height and (a) arsenic, (b) copper, (c) mercury, and (d) zinc.

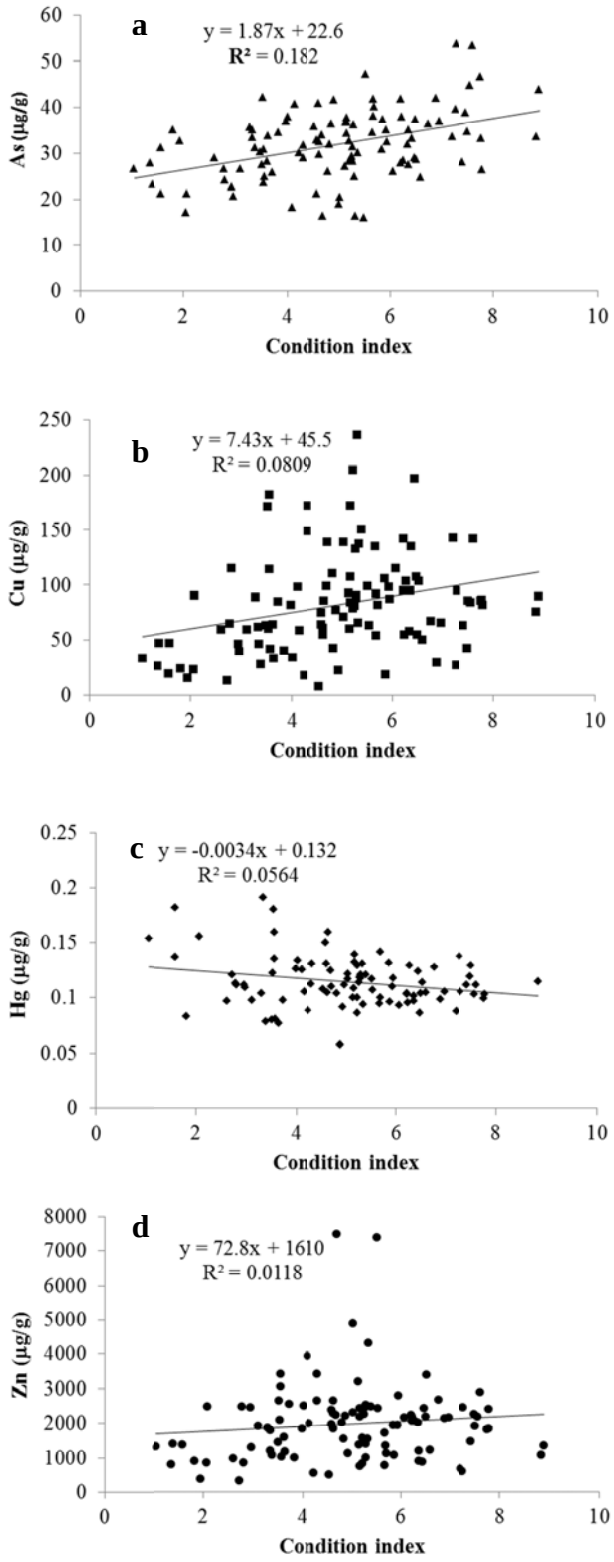


Figure 5. Correlations between condition index and (a) As, (b) Cu, (c) Hg, and (d) Zn concentrations in individual oysters.

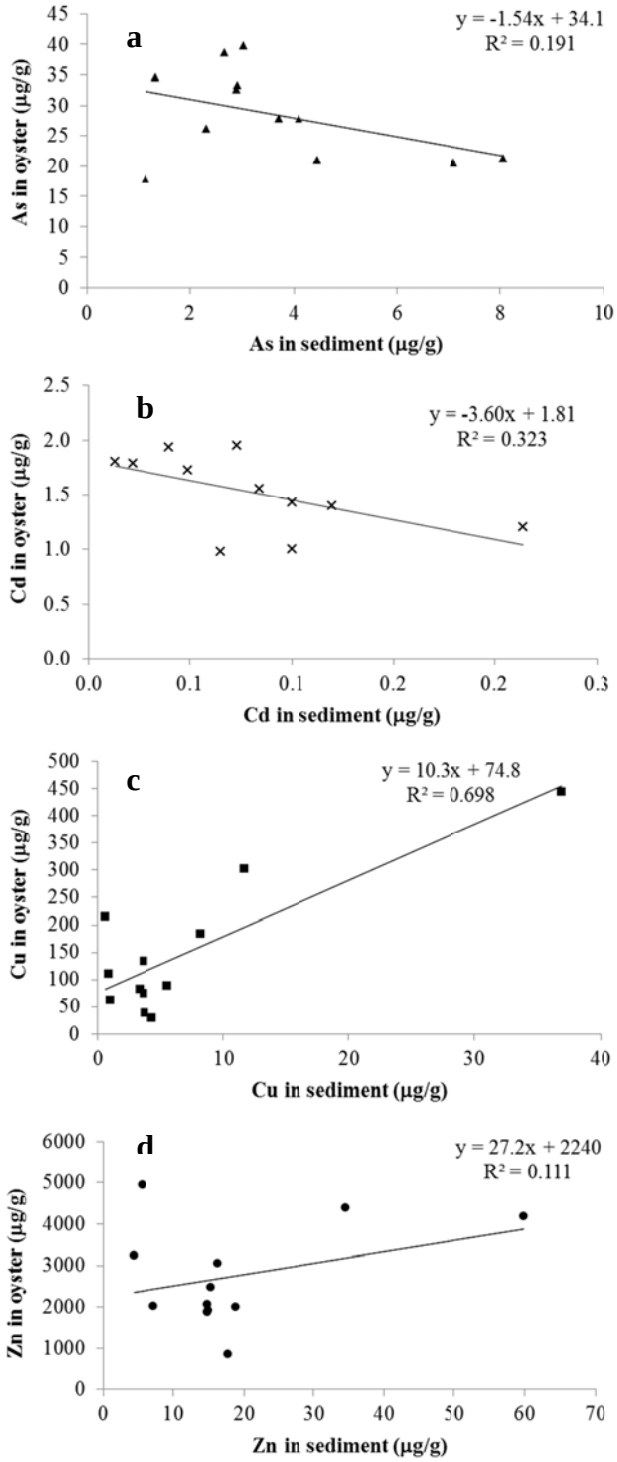


Figure 6. Correlations between metal concentrations in sediments and oyster tissues at sampling sites. Probabilities for correlations are (a) As, $p = 0.155$; (b) Cd, $p = 0.0680$ (c) Cu, $p = 0.000709$ (d) Zn, $p = 0.289$.

Organic contaminants in oysters and sediments

Polycyclic aromatic hydrocarbons

A series of 17 PAHs, listed in Table 1, was determined in composite oyster tissue samples (normally 20 oysters per composite) and adjacent sediments at 12 sites. Table A1 lists concentrations for individual PAHs as well as the sum of all 17 PAHs (ΣPAH), which are also summarized in Figure 7. Concentrations of ΣPAH ranged widely from 0.3-279 and 0.1-4960 ng g^{-1} in oysters and sediments, respectively. Oyster concentrations barely exceeded or were near the national median of 300 ng g^{-1} (Lauenstein et al., 2002) at only 3 sites (337, 293, 279 ng g^{-1} at LWF-3, LWF-4, and BC-H, respectively. Concentrations at the remaining sites were 4 to >10 times less than the national median.

PAH	TEL	PEL
	ng g^{-1} (ppb)	
Acenaphthene	6.71	88.9
Acenaphthylene	5.87	128
Anthracene	46.9	245
Benzo(a)anthracene	74.8	693
Benzo(a)pyrene	88.8	763
Benzo(b)fluoranthene	n/d	n/d
Benzo(g,h,i)perylene	n/d	n/d
Benzo(k)fluoranthene	n/d	n/d
Chrysene	108	846
Dibenzo(a,h)anthracene	6.22	135
Dibenzofuran	n/d	n/d
Fluoranthene	113	1494
Fluorene	21.2	144
Indeno(1,2,3-cd)pyrene	n/d	n/d
Naphthalene	34.6	391
Phenanthrene	86.7	544
Pyrene	153	1398
ΣPAH	1684	16770

Table 1. List of polycyclic aromatic hydrocarbons (PAHs) determined in this study. Also shown are criteria for the threshold effects level (TEL) and probably effects level (PEL) for 13 PAHs and for SPAH from MacDonald et al. (1996). n/d denotes value not determined. See text for definitions of TEL and PEL.

The toxicity status of the sediment was evaluated for each PAH and for ΣPAH by comparison to threshold effects level (TEL), defined as the concentration below which adverse effects rarely occur, and the probably effects level (PEL), defined as the concentration above which adverse effects can be expected to frequently occur (MacDonald et al., 1996). The values which have been developed for 12 individual PAHs and ΣPAH are shown in Table 1. Values of the TEL were exceeded for 10 PAHs and ΣPAH at site BC-H, for 7 PAHs and ΣPAH at site BH-3 and for 1 PAH at site BH-1. No values exceeded the PEL at any site. There were no correlations between individual PAH concentrations in sediments and oysters; this was greatly influenced by the large number of non-detectable values. There was no correlation between ΣPAH in sediments and oysters ($p > 0.2$).

Triclosan

Concentrations of triclosan were determined in composite oyster tissue samples and adjacent sediments at four sites: BC-H, HC-M, WC-M, and BH-1 (Figure 8). These results are the first paired oyster and sediment study of this commonly used personal care product. Levels of

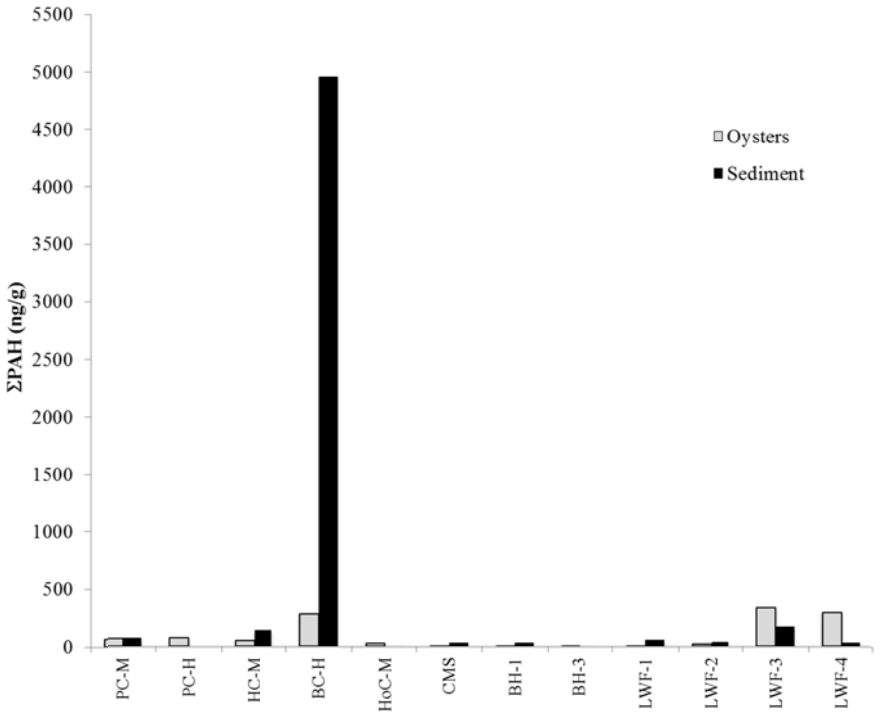


Figure 7. Concentrations of total PAHs (ΣPAH) in composite oyster tissue samples and adjacent sediments at sites in New Hanover and Brunswick counties.

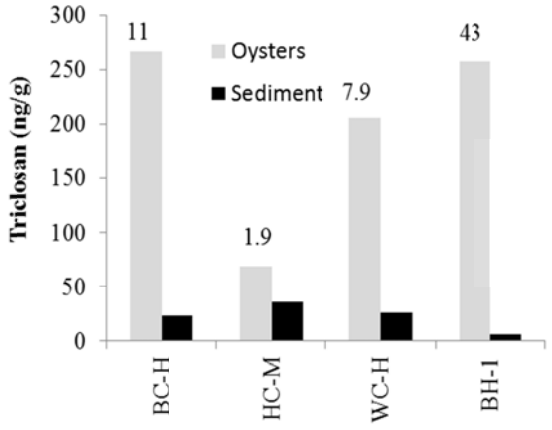


Figure 8. Concentrations of triclosan in composite oyster tissue samples and adjacent sediments. Values above the columns are the bioconcentration factors; i.e., the oyster concentration divided by the sediment concentration.

triclosan in oyster tissues and sediments ranged from 69-270 and 6.0-36 ng g^{-1}, respectively. The significance of correlation between sediment and oyster content could not be tested quantitatively because of the small sample size; however, qualitatively there is a trend of increasing oyster concentration with decreasing sediment triclosan content.

Although triclosan concentrations have been compiled for a variety of plants and invertebrates, including terrestrial and marine species (Amorim et al., 2010; Lyndall et al., 2010), to our knowledge we report the first concentrations of this contaminant in oyster tissues. Triclosan concentrations are consistently higher in tissues relative to the adjacent sediments. The ratio of these concentrations, referred to as the bioconcentration factor, ranges from 1.9 to 43, with the highest value occurring at the Bald Head harbor site, BH-1 (Figure 8).

DISCUSSION

Metals of concern

Table 2 lists the concentrations of metals found in oysters on the East and Gulf coasts of the United States in comparison to the results of this study. Of particular note are the values for the national medians from the NOAA National Status and Trends Program (Lauenstein et al., 2002) and the median for As in oysters from the southeastern US (Valette-Silver et al., 1999). These comparisons indicate that of all the metals in oyster tissues in New Hanover and Brunswick counties, As, Cu, and Zn are highest in comparison to other studies and to the national medians for at least some sites. The means and standard deviations of the 19 composite samples for these metals are 29.5 ± 7.1, 126 ± 104, and 2370 ± 1210 $\mu g\ g^{-1}$, respectively. Individual composites at the 19 sites all exceed the national median, and often the 85th percentile and SE US median, for As. Five sites exceeded the national median or 85th percentile for Cu, although the mean of the composite was lower than the national median, indicating localization of high values. Nine sites for Zn exceeded the national or 85th percentiles, and the mean of the composites was slightly above the national median. All other metals analyzed in the study were at or below national medians.

Previous studies have shown that there can be significant seasonal differences in oysters from a given site, with the highest tissue concentrations for many metals highest in summer relative to the other seasons. This may be due to the concentration of metals in the oyster tissue by a decrease in tissue glycogen content as a result of stresses due to spawning, temperature, salinity or other factors (Shumway, 1996). For example, Abbe et al. (2000) observed increases in concentration of up to twofold for Cu and up to fourfold for Cd in oysters in summer relative to other seasons in the Patuxent estuary. Jiann and Presley (1997) found differences of up to three- to four-fold higher concentrations of Ag, Cd, Cu, Pb, and Zn in summer compared other seasons in Galveston Bay oysters; little seasonal variability in As was observed. In contrast, Reidel et al. (1998) reported little variability in As concentrations in Patuxent estuary oysters.

In this study, some sites were sampled in July (HoC-M, PC-M, PC-H, HC-M, HC-H) or in early September (Lockwood Folly sites). However, generally these sites did not contain the highest concentrations of metals in the study. Thus, seasonal variability does not likely significantly affect the conclusions of this study.

Site	As	Cd	Cu	Hg	Pb	Ni	Ag	Zn	Reference
	Mean or median*	**Mean or median***	**Mean or median***	**Mean or median***	**Mean or median***	**Mean or median***	**Mean or median***	**Mean or median***	
Chesapeake Bay (Hog Island), NC	7.3								1
Cape Fear (Battery Island), NC	33								1
Winyah Bay (Lower bay), SC	46								1
Charleston Harbor (Ft. Johnson), SC	45								1
Charleston Harbor (Folly Island), SC	33								1
Sapelo Sound, GA	40								1
Patuxent estuary, MD	4.5								2
Charleston Harbor, SC	15.2								2
NC and SC	9.2	2.8	140*	0.10	0.51*	1.9*		2200*	3
Coastal waters, US	9.1*	2.0*	109*	0.11*	0.80*			1768*	4
Patuxent estuary, MD		13.3-14.0	115-130						5
Galveston Bay, TX	4.8-5.1	2.75-4.30	100-220		0.49-1.0		2.1-3.5	1440-2970	6
Patuxent estuary, MD		18.7-27.7	160-276				2.44-5.26		7
National median	9.2*	2.8*	140*	0.10*	0.51*	1.9*		2200*	3
National 85th percentile	16*	5.9*	290*	0.21*	0.82*	2.1*		4600*	3
SE US median	20.6*								1
New Hanover and Brunswick counties, NC	17.9-44.8	0.97-2.1	29-444	0.080-0.16	BD-0.56	BD-0.90	0.30-2.7	469-4950	This study

Table 2. Metal concentrations in oyster tissues in the mid-Atlantic and southeastern US. Concentrations all expressed in dry weights. References and notes: (1) Valette-Silver et al. (1999). Data for oysters. (2) Riedel and Valette-Silver (2002). Monthly means of oysters. (3) Lauenstein et al. (2002). Cu, Pb, Zn data for oysters only, other data for mussels and oysters. (4) O'Connor (1998).All data for 1996, Cu and Zn for oysters only, other metals mussels and oysters. (5) Abbe et al. (2000). Monthly means of oysters. Includes sites near power plant discharge water. (6) Jiann and Presley (1997). Range of seasonal means of oysters. (7) Riedel et al. (1998). Annual means for oysters from 1986-1996 for Cu and Ag, 1991-1996 for Cd. (This study). Ranges of means of composite samples of oysters.

Metal sources

Arsenic

Previous work on As in oysters and sediments has established that levels of this metal are higher in the southeastern US relative to the rest of the nation (Valette-Silver et al., 1999; Riedel and Valette-Silver, 2002). In North Carolina, elevated levels of As were detected in oysters collected off Battery Island at the Cape Fear mouth (Valette-Silver et al., 1999). There are several likely sources for the elevated As levels. First, the southeastern US from Virginia to Florida has extensive outcroppings of phosphorites, phosphate-rich biogenic sedimentary deposits. Arsenic is typically enriched in these deposits due to the chemical similarity between the phosphate (PO_4^{3-}) and arsenate (AsO_4^{3-}) ions. The relative importance of this source on As accumulation in southeastern oysters is difficult to assess; although phosphoritic sediments in the Southeast typically have higher than average As concentrations, oysters growing directly on such sediments do not normally have remarkably high As levels (Valette-Silver et al., 1999).

A second source of As is the historically widespread use of pesticides, fungicides, and herbicides containing predominantly inorganic As salts in the southeastern US. For example, in 1987 over 3.4 million kg (7.6 million lb) of these chemicals were applied in the drainage basins of Winyah Bay (SC), Albemarle-Pamlico Sound (NC) and the Cape Fear River (Pait et al., 1992 cited in Valette-Silver et al., 1999). Arsenic from these sources persists in soils and sediments long after application has ceased due to strong binding, particularly with Fe, Al, and Mn oxides and clay minerals abundant in most soils and sediments (Wauchope and McDowell, 1984; Goldberg, 2002). The large majority of As in soils, sediments, and plant materials is in the inorganic forms, arsenite and arsenate, with usually only a small fraction appearing as organic forms (Zheng et al., 2003). In contrast, a significant fraction of total As in marine organisms can be as organic forms (Le et al., 1994).

A third source of As and Cu (discussed further below) is pressure-treated lumber which has been manufactured since the 1930s using chromated copper arsenate (CCA). Such lumber has been widely used for coastal construction applications, including buildings, piers, and bulkheads. A number of studies have demonstrated that As and Cu can be greater in organisms and sediments and that toxicological effects on biota can be higher near areas containing CCA-treated wood such as bulkheads (Weis and Weis, 1992, 1993). However, metal concentrations in biota and sediments and their subsequent toxicological effects depend on a number of factors, including distance from the wood, its age, the fineness of adjacent sediments, and physical factors such as water flow (Weis et al., 1998).

Copper

Copper is ubiquitous in land-based and marine applications, including plumbing, wiring and electronic devices, agricultural chemicals, animal feed additives, and chemical manufacturing. As such Cu is one of the many metals with multiple sources that is considerably enriched in runoff from paved surfaces (Göbel et al., 2007), agricultural fields (Elrashidi et al., 2007) and concentrated animal operations (Burkholder et al., 2007). In developed coastal areas with many boating activities, a significant source of Cu is antifouling boat bottom paints, which are

designed to leach slowly into the water, releasing biocidal Cu ions. These paints may contain up to 70% by weight Cu. Numerous studies have found elevated concentrations of Cu in water, sediments, and biota near marinas (e.g., McGee et al., 1995; Schiff et al., 2004, 2007). In a previous study, we detected dissolved Cu at the Carolina Beach fishing charter docks which was about 10-fold higher than ambient Cu concentrations in the lower Cape Fear estuary (Larsen, 2010).

The probable effects of marinas as a source of Cu to oysters in southeastern North Carolina is reflected in the higher concentrations found in areas with relatively large numbers of recreational vessels. These areas include Bradley Creek and Whiskey Creek where composite Cu concentrations were above the national median (BC-M, WC-M) or 85th percentile (BC-H, WC-H). The Wrightsville Beach site (WB-NB) also exceeded the national median, but as the site was not adjacent to a marina, it is not clear if the major source is road runoff (the site was near a road bridge), boating activities, or other sources. The heads of the Bradley, Hewletts, and Whiskey creeks contained larger Cu concentrations than the mouths probably reflects trapping of Cu on the fine-grained sediments that tend to accumulate upstream (Renwick and Ashley, 1984).

Zinc

Zinc has numerous environmental sources, including industrial processes, rubber, plastics, paints, galvanized metal surfaces, and animal feed additives. A major source of Zn in road runoff is from the wearing of vehicle tires which contain about 1% by weight Zn (Davis et al., 2001; Councell et al., 2004). As for Cu, concentrated boating activities contribute Zn to coastal waters in the form of antifouling bottom paints and marine parts such as sacrificial anodes (Bird et al., 1996; Konstantinou and Albanis, 2004). Accordingly, the highest Zn concentrations were found in locations containing the highest density of recreational boats—Bradley, Hewletts, and Whiskey creeks. Bradley and Whiskey creek oysters significantly exceeded the national median and the head of Hewletts Creek exceeded the 85th percentile. Concentrations at the heads of these creeks, as well as Howe and Pages creeks, were higher than those at the mouth, consistent with Zn inputs from impervious surfaces in these watersheds. Previous work in the Cape Fear watershed has demonstrated the positive correlation between impervious cover and water quality indicators (Mallin et al., 2000).

Relationships between oyster physical parameters and metal concentrations

Metals vs. shell height

The lack of correlation between Cu and Zn concentrations and shell height in this study is contrary to the results of Birch et al. (2014), who found statistically significant positive relationships between shell height and levels of Cu and Zn in Sydney rock oysters, *Saccostrea glomerata*, in the highly impacted Sydney estuary, Australia. Previous studies of trace metals in oysters from the southeastern US did not report the relationship with shell height (Riedel et al., 1998; Valette-Silver et al., 1999; Lauenstein et al., 2002; Riedel and Valette-Silver, 2002).

Metals vs. condition index

The results of the present study for Cu are opposite to what was observed for *C. virginica* in Patuxent estuary, Maryland, in which there was a negative correlation between CI and metal concentration (Riedel et al., 1998). Avery et al. (1996) found a negative correlation between CI and Hg content in *S. glomerata* at impacted regions in New South Wales. Rebelo et al. (2005) found no correlations between CI and either Cu or Zn in the mangrove oyster, *C. rhizophorae*, and Páez-Osuna et al. (1995) found no relationships between CI and a number of metals including Cu and Zn in *C. iridescens*. A lack of consistent relationships between metal levels and CI is not surprising, since CI is dependent on a large variety of different factors, including physical conditions (e.g., temperature, salinity, tidal exposure), biological factors (e.g. parasites such as pea crabs, Dermo, MSX), and biochemical factors (nutrient availability, exposure to organics and metals) (Gold-Bouchot et al., 1995; Avery et al., 1996; Rice and Rheault, 1996; Mercado-Silva, 2005). Thus, CI by itself is not likely to be a valid indicator of metal stress on oysters.

Relationships between oyster and sediment metal concentrations

Previous studies that have examined the relationship between metal concentrations in oysters and nearby sediments do not demonstrate consistent patterns across different environments. Valette-Silver et al. (1999) found a positive correlation (R^2 of 0.23-0.39) between As in sediments and mollusk (oyster and mussel) tissues throughout the southeastern U.S. with most of the correlation driven by a relatively few data points representing tissues with high concentrations. Vázquez-Sauceda et al. (2011) found positive correlations between Cd and Pb in sediments and *C. virginica* tissues from San Andreas Lagoon, Mexico, but no relationship with Zn. No correlations were found between sediment and *S. glomerata* concentrations of Cu and Zn in Sydney estuary, although the relationship for Zn became significant and that for Cu improved when the metal content of the suspended particulate material (SPM) was considered (Birch et al. (2014). The latter study did show a strong positive correlation for Pb in tissues for both surface sediments and SPM. Chaharlang et al. (2012) found strong correlations between Cd, Cu and Zn in tissues of the hooded oyster (*S. cucullata*) from the Persian Gulf and ambient fine sediment (<62.5 µm), but not for Pb.

The lack of consistent and strong correlations in the present study and in previous work suggests that concentrations of metals in ambient surface sediments are not robust predictors of metal levels in oyster tissues. This appears to be true even when metal concentrations of SPM are considered, even in very highly impacted environments such as the Sydney estuary, Australia, where mean tissue metal (Cu, Pb, Zn) concentrations are up to 5-10 times greater than those found in the present study, with much higher sediment metal levels (Birch et al., 2014). Since oysters are filter feeders, the metals they accumulate will be dominated by those present in the suspended particulate matter that is accessible to them. In addition, active bioaccumulation of certain metals and variable rates of depuration of different metals will affect the relative levels of tissue metals in comparison to sediment and suspended particulate matter levels (Ke and Wang, 2001).

Organic contaminants

Distributions and sources of PAHs

The presence of PAHs in coastal sediments in southeastern North Carolina is expected because of the multiple sources of these compounds, which are derived from burned and unburned fossil fuels. Such sources include atmospheric deposition from combustion processes (coal, wood, petroleum-derived fuels), fuel spills, and lubricants. Waterbodies in particular receive PAHs from runoff from paved surfaces, creosote-treated lumber, and boating and shipping activities. Thus, sediments in highly urbanized coastal and marine areas are typically very high in PAHs, which consequently bioaccumulate in a wide variety of biota (Meador et al., 1995; Lima et al., 2005).

Sediments at 2 of the 12 sites analyzed in this study contained concentrations of PAHs (both individual PAHs and ΣPAH) at or above the TEL, a criterion above which adverse effects might start to be observed. These sites, which can be classified as moderately impacted, have very different physical settings and likely different sources for the PAHs. Site BC-H is in the upper tidal portion of Bradley Creek, proximal to both active marinas and a heavily traveled roadway between Wilmington and Wrightsville Beach and points north (Oleander Dr./Route 76). The Bradley Creek watershed is one of the largest and most highly developed in New Hanover County, with a population of 16,500, 23% impervious surface, and chronic water quality problems as reflected by low oxygen levels and high fecal coliform bacteria counts (Mallin et al., 2013). In contrast, the BH-3 site, located on Bald Head Island, a lightly populated community with few motor vehicles, had an unexpected 8 TEL exceedances for 7 PAHs and ΣPAH. This site is within the interior of the island, not close to boating facilities. Bald Head Creek is poorly flushed with fine-grained sediments. High fecal coliform counts have been reported in this area; these sources are being investigated by the Bald Head Island Conservancy (https://www.bhic.org/salt-marsh).

Inferences about the source of PAHs can be made using diagnostic ratios of the PAHs in oysters and sediments for which data are available. The ratios were anthracene divided by the sum of anthracene plus phenanthrene (An/An + Ph) and fluoranthene divided by the sum of fluoranthene plus pyrene (Fl/Fl+Py) (e.g., Westerholm and Li, 1994; Tobiszewski and Namieśnik, 2012). Table 3 presents these diagnostic ratios as well as the ranges for known pyrogenic and petrogenic origins. Pyrogenic refers to combustion-derived whereas petrogenic refers to non-combusted fossil fuel origins such as oil or fuel spills. For example, sediments collected from the Frasier River, Canada had Fl/Fl+Py values >0.5 suggesting combustion of wood or other biomass (Yunker et al., 2002). As a comparison, sediments from San Francisco Bay had a Fl/Fl+Py mean range of 0.43-0.45, indicating dominance by petrogenic sources of PAHs (Oros and Ross, 2004). PAH signatures from oysters and sediment in southeastern North Carolina clearly reflect pyrogenic sources from the combustion of fossil fuels that are likely delivered to coastal waters by boating activities and road runoff.

	An/(An+Ph)		Fl/(Fl+Py)	
Pyrogenic sources	>0.1		>0.4	
Petrogenic sources	<0.1		<0.4	
Site	Oyster	Sediment	Oyster	Sediment
BC-H	0.4	0.3	0.5	0.5
CMS		0.7		0.5
HC-M		0.6	0.5	0.6
HoC-M		0.6		0.6
PC-H	0.3	0.5	0.5	0.5
PC-M	0.5	0.7	0.3	0.6
BH-1		0.6		0.2
BH-3				
LWF-1		0.4		0.5
LWF-2		0.3		0.5
LWF-3	0.4	0.5		0.6
LWF-4		0.4		0.5

Table 3. The interpretation of ratios of An/(An+Ph) and Fl/(Fl+Py) as source indicators of PAHs (top of table) and calculated ratios in composite oyster tissues and adjacent sediments at selected sites. An = anthracene; Ph = phenanthrene; Fl = fluoranthene; Py = pyrene.

There are few published data on PAH concentrations in North Carolina sediments and oysters with which to compare our data. However, there are a number of studies from South Carolina and national and regional results with which to compare our results.

Lauenstein et al. (2002) summarized the results of the NOAA Status and Trends ("Mussel Watch") project at 11 stations in the Carolinas. This included 7 stations in North Carolina, only one of which was in southeastern North Carolina, at Battery Island in the lower Cape Fear estuary. Concentrations of ΣPAH at most of these stations, including Battery Island, were near the national median. A few stations, including 2 in the heavily impacted Charleston Harbor, and 1 site at the Pivers Island bridge in Beaufort, NC had relatively high concentrations of PAHs. The high Charleston Harbor concentrations found in oysters (Lauenstein et al., 2002) and sediments (Kucklick et al., 1997) is consistent with impacts due to boating and shipping activities and extensive urbanization of the watershed. High concentrations at the Pivers Island bridge was attributed to proximity to a roadway (Lauenstein et al., 2002) and is consistent with the high concentrations of PAHs near the roadway bridge near site BC-H in the present study, a consequence of the more modest urbanization in the greater Wilmington area.

Table 4 shows a comparison of ΣPAH results in sediments and oysters from this study to other results from the southeastern US and nationally. Beyond the two somewhat elevated sediment concentrations in this study (1765 and 4960 ng g^{-1} at BH-3 and BC-M, respectively), the remaining samples (0.067-171 ng g^{-1}) are at the low range compared to existing results. Concentrations in oysters are at the lower range of all measurements, lower than national ranges and comparable to environments such as North Inlet, SC, which is only moderately impacted. As reported in other studies, controlling factors for sediment accumulation of PAHs often include grain size and concentrations of organic carbon, which has an affinity for hydrophobic PAHs. The most dominant factor, however, is proximity to sources of road runoff or other pyrogenic PAH combustion sources (Kucklick et al., 1997; Lauenstein et al., 2002). This seems to be the case for the relatively few elevated PAH values in this study.

Site	Range in sediments (ng g^{-1})	Range in oysters (ng g^{-1})	Reference and notes
3 coastal marinas, SC	496 – 8360	---	1
Murrells Inlet, SC	14 – 2578	56 - 1968	2
North Inlet, SC	9 – 308 (5847)	15 -172	2
North Edisto estuary, SC	33 – 221		3
Charleston Harbor, SC	34 - 9630		3
Winyah Bay, SC	157 – 1990		3
East, West, Gulf coasts, US		192 - 503	4
East Coast, US		<100 - 2700	5
New Hanover and Brunswick counties, NC	14.5 - 4960	0.3 - 337	This study

Table 4. Total PAH (SPAH) concentrations in sediments and oyster tissues in the US with a focus on the Southeast. Concentrations all expressed in dry weights. References and notes: (1) Marcus and Stokes (1985). PAHs in oysters were reported on a wet weight basis and are not shown here. (2) Sanders (1995). Value in parentheses for sediments in North Inlet was one high value out of 31 reported concentrations. (3) Kucklick et al. (1997). (4) O'Connor (1998). Range of annual medians in mollusks (mussels and oysters) from 1988-1996 Mussel Watch results; 97-191 sites per year. (5) Kim et al. (2008) Multiple sites along East Coast, US.

Polycyclic aromatic hydrocarbons are aromatic compounds composed of 2 to 6 rings. For example, naphthalene, anthracene, fluoranthene, benzo(a)pyrene, and benzo(g,h,i)perylene contain 2, 3, 4, 5, and 6 rings, respectively. In general, PAHs with fewer number of rings are slightly more soluble (i.e., less hydrophobic) and more easily taken up by biota than are PAHs with more rings. This is consistent with the pattern of 2 and 3-ringed PAHs being relatively more abundant in oyster tissue, and PAHs of higher molecular weight (larger rings) dominating in sediments (Figure 9). These patterns are consistent with those found in other studies of oyster-sediment PAH partitioning (Sanders, 1995).

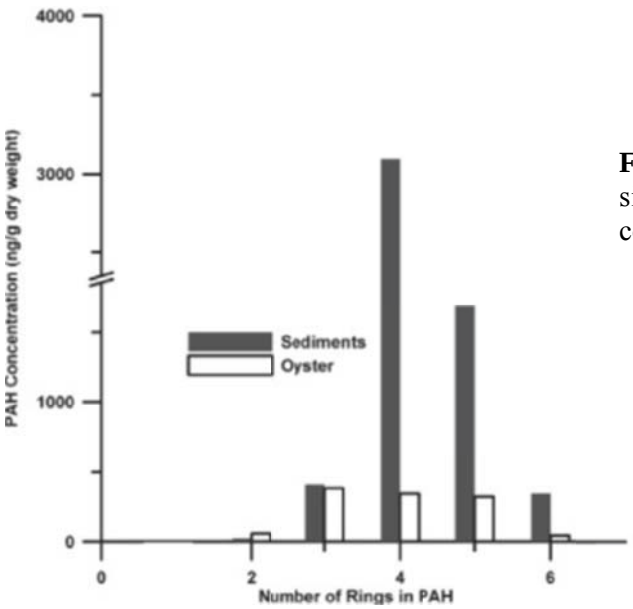


Figure 9. Distribution of PAHs of differing ring sizes in sediments and composite oyster samples collected in this study.

Distribution and sources of triclosan

Sediment concentrations of triclosan found in this study, ranging from 6.0 to 36 ng g^{-1}, are comparable to or at the low ranges of those reported previously in surface sediments, most of which are known to be impacted by inputs from triclosan sources such as wastewater treatment plants (Table 5).

Sample Site	Triclosan (ng g^{-1})	References and notes
Almería, SE Spain	0.27 – 131	1
Virginia, US	10 – 160	2
Chesapeake Bay, MD and Jamaica Bay, NY	<10 – 560	3
NW Spain	<0.40 – 36	4
Sweden	<2 – 25	5
Lower Hudson River, NY	9.0 – 37	6

Table 5. Concentrations of triclosan (dry weight basis) in surface sediments reported in various studies. References: (1) Agüera et al. (2003). Marine sediments adjacent to WWTP. (2) Hale et al. 2000. Higher concentration found near decommissioned STP. (3) Miller et al. (2008). Sediments near WWTP. (4) Morales et al. (2005). (5) Remberger et al. (2002). (6) Wilson et al. (2008).

Concentrations of triclosan found in biological samples including fish and bottlenose dolphins range from <0.033 to 31 ng/g (Lyndall et al., 2010). Previous studies on levels in fish and marine mammals indicate that triclosan bioaccumulates and persists in tissues (Valters et al., 2005; Fair et al., 2009). Its relatively high octanol-water coefficient (log K_{ow} = 4.67) suggests a lipophilic

nature that enhances accumulation in fatty tissues (Geens et al., 2012). Triclosan levels in oyster tissues of this study ranged from 69 to 267 ng g^{-1}, about 2-9 times greater than those found previously in biological samples. Based on comparison with bioaccumulation of other relatively lipophilic contaminants, such as polychlorinated biphenyls (PCBs), it is likely that triclosan is largely associated with the lipid-rich components of oyster tissue, although future studies are needed to confirm this.

The HC-M and BC-H sites are closest to the most urbanized watersheds which have experienced numerous sewage spills (City of Wilmington, 2012). Thus, it was expected that these locations would have the highest concentration of contaminants such as triclosan. As expected, the concentrations of triclosan in sediment from these sites were the highest in the study. Detectable concentrations at the Whiskey Creek site (WC-H) due to urbanization were expected. As expected, the Bald Head Island site (BH-1) is the least developed and had the lowest sediment concentration. However, the oyster tissue samples did not all follow the expected pattern of more urbanized areas having larger triclosan concentrations. Although BC-H oysters showed the highest triclosan concentrations, the second-highest levels found at BH-1 were unexpected given that it is a lightly developed area. One possibility is that triclosan releases from upstream sites in the Cape Fear estuary are accumulated over time at the mouth. Another possibility is an undetected source of sewage or input from boating activities near the Bald Head Harbor.

Triclosan can be transformed during wastewater treatment and in the environment through a number of chemical, microbial, and photochemical pathways (Tixier et al., 2002; Balmer et al., 2004; Suarez et al., 2007). Some photochemical products are dioxins, which have relatively high toxicity and bioaccumulation potential in biota (Latch et al., 2003). In wastewater treatment, a common metabolite is methyl triclosan which is formed by methylating bacteria and is more lipophilic and environmentally persistent than triclosan (Coogan et al. 2007). Neither methyl triclosan nor dioxin photolysis products of triclosan were observed in oyster tissues and sediment samples in this study.

CONCLUSIONS AND RECOMMENDATIONS

Metals and organic contaminants in oyster tissues in New Hanover and Brunswick counties

The metals of greatest concern is oyster tissues in the region are As, Cu, and Zn, because they occur at relatively high concentrations at some or all sample locations compared to national median levels. Mercury is a contaminant of global concern because of sources that are distributed worldwide (e.g., coal burning, smelting, cement manufacture) and its tendency to convert in coastal environments to methylmercury, a bioaccumulative neurotoxin. However, average Hg concentrations in oysters in the study area are at or near the national median, a level which appears to be a minimal risk to humans who consume oysters (see below). The levels of As in oysters consistently exceeds national median and 85th percentile values. This is consistent with the elevated As concentrations found previously in oysters and sediments from the southeastern US. This large background level is likely associated with relatively high concentrations in phosphorite-rich bedrock, soils, and sediments, and historical inputs from pesticide applications in the Cape Fear watershed.

Elevations of Cu and Zn near urbanized areas and marinas likely reflect inputs from boating activities, particularly antifouling paints and sacrificial anodes, as well as stormwater runoff. Treated lumber, as used for bulkheads and piers, may be localized sources of As and Cu. Elevations of PAHs were found in sediments close to marinas and roadways, with the most impacted site at Bradley Creek (BC-H) near a heavily used roadway. Unexpected elevations of PAHs occurred in a salt marsh site (BH-3) in lightly populated Bald Head Island; the source of this elevation is not known and merits further study. The BC-H and BH-3 sites showed some PAH concentrations in sediments that exceeded threshold effects levels at which some toxic effects might begin to be observed. Oyster tissues at all sites were all near or below national median concentrations. PAHs in oyster tissues were dominated by smaller molecular weight species, consistent with their relatively greater solubility and bioavailability.

Triclosan was detected at all 4 paired oyster-sediment sites in this study, reflecting the wide-ranging usage of this common antibacterial compound in a variety of consumer products. Levels detected were well within ranges found in sediments and organisms in previous studies; however, our results are the first reported for oyster tissues.

The results of our study indicate that most contaminant effects on oyster tissues, except for As, are related to localized sources of pollution, such as marinas, roadways, and heavily developed areas. Areas undergoing oyster restoration in Lockwood Folly are likely to yield oyster tissues with unexceptional levels of metals and organic contaminants, reflecting the relatively low development of its borders. Likewise, oyster reefs away from intense development are more likely to yield oysters with lower contaminant levels than those near highly urbanized areas. Oyster restoration efforts near impacted areas should improve water quality because of filtration of particulates and associated contaminants by filter feeding, even though contaminant levels (more likely of fecal coliform rather than metals or organics) would restrict consumption.

Possible human health effects from potential contaminants in oyster tissue

It is beyond the scope of this project to provide an extensive analysis of potential human health effects from ingestion of oysters with contaminant loads similar to those found in this study. However, we present some simple calculations that suggest that normal ingestion of oysters from southeastern North Carolina is unlikely to exceed accepted dietary recommendations. We limit our discussion to the four metals at greatest concentration or of highest national concern: As, Cu, Hg, and Zn. There are no guidelines for PAH or triclosan ingestion. Menzie et al. (1992) conclude that PAH intake from food is likely to be the most important source for nonsmokers. The locations at which the highest concentrations of PAHs, triclosan, and metals are found in this study are nearest highly impacted areas such as marinas or near urbanized zones. These areas are normally closed to shellfishing (usually due to high fecal coliform counts), so harvesting of oysters for consumption is not legally allowed.

For our calculations we assume an individual of 60 kg (132.3 lb) and used available national or international human intake criteria. All calculations assume ingestion of 12 oysters each of 0.5 g dry weight with conservative (biased toward the high end of the range) estimates of metal concentrations in the oyster tissue.

The maximum suggested intake for As (assuming all As is in the inorganic forms) is 3 μg kg^{-1} (WHO, 2010). Ingestion of 12 oysters containing 30 μg g^{-1} yields 180 μg total which is the recommended value of 3 μg kg^{-1}. Daily consumption of this many oysters is highly unlikely for most individuals. Copper is a necessary nutrient for humans; the recommended daily allowance (RDA) is 900 μg day^{-1} for males above the age of 19 (IOM, 1998). Consumption of 12 oysters weighing 0.5 g each at a concentration of 100 μg g^{-1} gives a total dose of 600 μg, a value below the RDA. The conservative criterion of 1.6 μg kg^{-1} $week^{-1}$ of mercury (as methylmercury) (UN FAO/WHO, 2003), or 96 μg $week^{-1}$ for a 60-kg nursing female is well above the 0.6 μg provided by a weekly ingestion of 12 oysters each containing 0.1 μg g^{-1} Hg. Zinc is another required nutrient, with a RDA of 11 mg day^{-1} for males aged 14 and above (IOM, 1998). Ingestion of 12 oysters each containing 2500 μg g^{-1} yields 15 mg which exceeds the RDA, but when averaged over a week, can be considered only a reasonable dietary source of Zn.

OUTREACH AND EDUCATIONAL ACTIVITIES

The large majority of the work presented here was performed by undergraduate students in the Department of Chemistry and Biochemistry at UNCW. Three students (Savannah Sherard, Lauren Kipp, and Mary Steagall) performed honors projects and numerous students (including Aaron Vick, Jessica Ramirez, Karen Dodson, Melissa Lyles, Hannah Liberatore, Graham Collier, Paula McConnell, Christina DeFrancesco, Amy Barbera, and Karen Dodson) performed research as directed individual study projects. We are grateful for the tremendous assistance of these students.

We are still being assisted by graduate student Kaitlyn Costin to finish the website that will display all sample sites and data collected as part of this study. This component has been delayed due to time conflicts.

Drs. Mead and Skrabal have discussed their results in venues such as the Tidal Creeks Symposium held in Wilmington, NC, in December 2013. Dr. Skrabal has discussed the oyster project to high school students attending the UNCW Marine Quest summer program in 2013 and 2014.

We will be preparing a manuscript based on the results of this study for publication in a peer-reviewed journal.

Literature cited:

Abbe, G.R., Riedel, G.F., and Sanders, J.G., 2000. Factors that influence the accumulation of copper and cadmium by transplanted eastern oysters (Crassostrea virginica) in the Patuxent River, Maryland. *Marine Environmental Research* 49, 377-396.

Agüera, A., Fernández-Alba, A.R., Piedra, L., Mézcua, M., Jose Gómez, M. 2003. Evaluation of triclosan and biphenyl in marine sediments and urban wastewaters by pressurized liquid extraction and solid phase extraction followed by gas chromatography mass spectrometry and liquid chromatography mass spectrometry. *Analytica Chimica Acta* 480, 193-205.

Amorim, M.J.B., Oliveira, E., Soares, A.M.V.M., and Scott-Fordsman, J.J., 2010. Predicted no effect concentration (PNEC) for triclosan to terrestrial species (invertebrates and plants). *Environment International* 36, 338-343.

Avery, E.L., Dunstan, R.H., and Nell, A., 1996. The detection of pollutant impact in marine environments: condition index, oxidative DNA damage, and their associations with metal bioaccumulation in the Sydney Rock Oyster *Saccostrea commercialis*. Archives of Environmental Contamination and Toxicology 31, 192-198.

Balmer , M.E., Poiger, T., Droz, C., Romanin, K., Bergqvist, P.-A., Müller, M.D., and Buser, H.-R., 2004. Occurrence of methyl triclosan, a transformation product of the bactericide triclosan, in fish from various lakes in Switzerland. Environmental Science and Technology 38, 390-395.

Bat, L. and Raffaelli, D., 1998. Sediment toxicity testing: a bioassay approach using the amphipod *Corophium volutator* and the polychaete *Arenicola marina*. *Journal of Experimental Marine Biology and Ecology*, 226, 217-239.

Birch, G.G.,Melwani, a., Lee, J.H., and Apostolatos, C., 2014. The discrepancy in concentration of metals (Cu, Pb and Zn) in oyster tissue (Saccostrea glomerata) and ambient bottom sediment (Sydney estuary, Australia) *Marine Pollution Bulletin* 80, 263–274.

Bird, P., Comber, S.D.W.,Gardner, m.J., and Ravenscroft, J.E., 1996. Zinc inputs to coastal waters from sacrificial anodes. *Science of the Total Environment* 181, 257-264.

Burkhardt, B.G. 2009. *Trace Metals in Tidal Creeks Sediments: The Effects of Anthropogenic Inputs, Wilmington, NC.* B.S. honors thesis, UNC Wilmington.

Burkholder, J., Libra, B., Weyer, P., Heathcote, S., Kolpin, d., Thorne, P.S., and Wichman, M., 2007. Impacts of waste from concentrated animal feeding operations on water quality. *Environmental Health Perspectives* 115, 308–312.

Chaharlang, B.H., Bakhtiari, A.R., and Yavari, V., 2012. Assessment of cadmium, copper, lead and zinc contamination using oysters (*Saccostrea cucullata*) as biomonitors on the coast of the Persian Gulf, Iran. *Bulletin of Environmental Contamination and Toxicology* 88, 956-961.

Chau, W.C., Wu, J., and Cai, Z. 2008. Investigation of levels and fate of triclosan in environmental waters from the analysis of gas chromatography coupled with ion trap mass spectrometry. *Chemosphere* 73, S13-S17.

City of Wilmington, 2012. Bradley and Hewletts creeks watershed restoration plan. Wilmington, NC. Available at: *http://www.wilmingtonnc.gov/Portals/0/documents/Public%20Services/Stormwater/G2B%20final%20plan%202012_August.pdf*.

Comber, S.D.W., Gardner, M.J., and Boxall, A.B.A., 2002. Survey of four marine antifoulant constituents (copper, zinc, diuron and Irgarol 1051) in two UK estuaries. *Journal of Environmental Monitoring* 4, 417-425.

Coogan, M.A., Edziyie, R.E., La Point, T.W., and Venables, B.J., 2007. Algal bioaccumulation of triclocarban, triclosan, and methyl-triclosan in a North Texas wastewater treatment plant receiving stream. *Chemosphere* 67, 1911-1918.

Couch, L.T., Posey, M.H., and Alphin, T.D. 2010. Environmental quality and oyster disease (abstract). 39th Benthic Ecology meeting, Wilmington, NC.

Councell , T.B., Duckenfield. K.U., Landa , E.R., and Callender, E., 2004. Tire-wear particles as a source of zinc to the environment. *Environmental Science and Technology* 38, 4206-4214.

Daskalakis, K.D., 1996. Variability of metal concentrations in oyster tissue and implications to biomonitoring. *Marine Pollution Bulletin* 32, 794-801.

Davis, A.P., Shokouhian, M., and Ni, S. 2001. Loading estimates of lead, copper, cadmium, and zinc in urban runoff from specific sources. *Chemosphere* 44, 997-1009.

DeLorenzo, M.E., Keller, J.M. Arthur, C.D., Finnegan, M.C., Harper, H.E., Winder, V.L., and Zdankiewicz, D.L. 2008. Toxicity of the antimicrobial compound triclosan and formation of the metabolite methyl-triclosan in estuarine systems. *Environmental Toxicology* 23, 224-232,

Dunning, H.N., Moore, J.W., Bieber, H., and Williams, R.B. 1960. Porphyrin, nickel, vanadium, and nitrogen in petroleum. *Journal of Chemical and Engineering Data* 5, 546-549.

Elrashidi, M. A., Hammer, D., Fares, A., Seybold, C.A., Ferguson, R., and Peaslee, S. D., 2007. Loss of heavy metals by runoff from agricultural watersheds. *Soil Science* 172, 876-894.

Fair, P.A., Lee, H.-B., Adams, J., Darling,C., Pacepavicius, G., Alaee, M., Bossart, G.D., Henry, N., and Muir, D., 2009. Occurrence of triclosan in plasma of wild Atlantic bottlenose dolphins (*Tursiops truncatus*) and in their environment. *Environmental Pollution* 157, 2248-2254.

Farrington, J.W., Goldberg, E.D., Risebrough, R.W., Martin, J.H., and Bowen, V.T. 1983. U.S. "Mussel Watch" 1976-1978: An overview of the trace-metal, DDE, PCB, hydrocarbon and artificial radionuclide data. *Environmental Science and Technology* 17, 490-496.

Geens, T., Neels, H., and Covac, A. 2012. Distribution of bisphenol-A, triclosan and n-nonylphenol in human adipose tissue, liver and brain. *Chemosphere* 87, 796-802.

Göbel, P., Dierkes, C., and Coldewey, W.G., 2007. Storm water runoff concentration matrix for urban areas. *Journal of Contaminant Hydrology* 91, 26–42.

Gold-Bouchot, G., Simá-Alvarez, R., Zapata-Pérez, O., and Güemez-Ricalde, J., 1995. Histopathological effects of petroleum hydrocarbons and heavy metals on the American oyster (Crassostrea virginica) from Tabasco, Mexico. *Marine Pollution Bulletin* 31, 439–445.

Goldberg, S., 2002. Competitive adsorption of arsenate and arsenite on oxides and clay minerals. *Soil Science Society of America Journal* 66, 413–421.

Grabowski, J.H. and, Peterson, C.H. 2007. Restoring oyster reefs to recover ecosystem services. In: Pages 281–298 in Cuddington, K, Byers, J.E., Wilson, W.G., and Hastings, A., (eds.) Ecosystem Engineers: Plants to Protists. p. 281-298. Elsevier.

Grabowski, J.H., Brumbaugh, R.D., Conrad, R.F., Keeler, A.G., Opaluch, J.J., Peterson, C.H., Piehler, M.F., Powers, S.P., and Smyth, A.R.. 2012. Economic valuation of ecosystem services provided by oyster reefs. *BioScience* 62, 900-909.

Guo, L, Santschi, P.H., and Ray, S.M. 2002. Metal partitioning between colloidal and dissolved phases and its relation with bioavailability to American oysters. *Marine Environmental Research* 54, 49-64.

Hale, R.C., Smith, C.L., de Fur, P.O., Harvey, E., Bush, E.O., La Guardia, M.J., and Vadas, G.G., onylphenols in sediments and effluents associated with diverse wastewater outfalls. *Environmental Chemistry and Toxicology* 19, 946-952.

Holland, A.F., Sangera, D.M., Gawle, C.P., Lerberg, S.B., Santiagoa, M.S., Riekerk, G.H.M., Zimmerman, L.E., and Scott, G.I. 2004. Linkages between tidal creek ecosystems and the landscape and demographic attributes of their watersheds. *Journal of Experimental Marine Biology and Ecology* 298, 151– 178.

Hopkins, A. E. 1949. Determination of condition of oysters. *Science* 110,567-568.

Hyland, J.L., Balthis, W.L., Hackney, C.T., and Posey, M. 2000. Sediment quality of North Carolina estuaries: an integrative assessment of sediment contamination, toxicity, and condition of benthic fauna. *Journal of Aquatic Ecosystem Stress and Recovery* 8, 107-124.

IOM (Institute of Medicine), 1998. Dietary reference intakes: A risk assessment model for establishing upper intake levels for nutrients. National Academies Press, Washington, DC.

Jiann, K.T. and Presley, B.J., 1997. Variations in trace metal concentrations in American oysters (*Crassostrea virginica*) collected from Galveston Bay, Texas. *Estuaries* 20, 710-724.

Ke,. C. and Wang, W.-X., 2001. Bioaccumulation of Cd, Se, and Zn in an estuarine oyster (*Crassostrea rivularis*) and a coastal oyster (*Saccostrea glomerata*).

Kim, Y., Powell, E. N., Wade, T.L., Presley, B.J. 2008. Relationship of parasites and pathologies to contaminant body burden in sentinel bivalves: NOAA status and trends 'mussel watch' program. *Marine Environmental Research* 65, 101-127

Konstantinou, I.K. and Albanis, T.A., 2004. Worldwide occurrence and effects of antifouling paint booster biocides in the aquatic environment: a review. *Environment International* 30, 235-248.

Kucklick, J.R., Sivertsen, S.K., Sanders, M., and Scott, G.I., 1997. Factors influencing polycyclic aromatic hydrocarbon distributions in South Carolina estuarine sediments. *Journal of Experimental Marine Biology and Ecology* 213, 13–29.

Landrum, P.F., Lotufo, G.R., Gossiaux, D.C., Gedeon, M.L., and Lee, J.-H., 2003. Bioaccumulation and critical body residue of PAHs in the amphipod, Diporeia spp.: additional evidence to support toxicity additivity for PAH mixtures. *Chemosphere* 51, 481-489.

Lauenstein, G.G., Cantillo, A.Y., and O'Connor, T. P., 2002. The status and trends of trace element and organic contaminants in oysters, Crassostrea virginica, in the waters of the Carolinas, USA. *Science of the Total Environment* 285, 79-87.

Larsen, L.A., 2010. The photolytic release of dissolved copper from resuspended sediments in coastal environments. M.S. thesis, University of North Carolina Wilmington, Wilmington, NC.

Latch, D.E., Packer, J.L, Arnold, W.A., and McNeill, K., 2003. Photochemical conversion of triclosan to 2,8-dichlorodibenzo-p-dioxin in aqueous solution. *Journal of Photochemistry and Photobiology*: A. Chemistry 158, 63-66.

Lawrence, D.R, and Scott, G.I. 1982. The determination and use of condition index in oysters. Estuaries 5, 23-27.

Le, X.-C., Cullen, W.R., and Reimer, K.J., 1994. Speciation of arsenic compounds by HPLC with hydride generation atomic absorption spectrometry and inductively coupled plasma mass spectrometry detection. *Talanta* 41, 495-502.

Le Dréau, Y., Doumenq, P., Lamontagne, J., Kister, J., and Mille, G. 2000. PAHs in sediments contaminated by the Haven oil spill (Genoese Bay). *Polycyclic Aromatic Compounds* 20, 111-122.

Lima, A.L., Farrington, J.W., and Reddy, C.M., 2005. Combustion-derived polycyclic aromatic hydrocarbons in the environment—A review. *Environmental Forensics* 6, 109-131

Lockwood Folly River Water Quality Strategy. 2007. Report to the Brunswick County Board of Commissioners, Lockwood Folly Watershed Roundtable (available at *http://www.nceep.net/services/lwps/Lockwood/Lockwoods%20Folly%20DA-TM%20Report%20Appendix%20D%20Final%2006-26-07.pdf*)

Lyndall, J., Fuchsman, P., Bock, M., Barber, T., Lauren, D., Leigh, K., Perruchon, E., and Capdevielle, M., 2010. Probabilistic risk evaluation for triclosan in surface water, sediments, and aquatic biota tissues. *Integrated Environmental Assessment and Management* 6, 419-440.

MacDonald, D.R., Carr, R.S., Calder, F.D., Long, E.R., and Ingersoll, C.G., 1996. Development and evaluation of sediment quality guidelines for Florida coastal waters. *Ecotoxicology* 5, 253-278.

MacPherson, T.A. Cahoon, L.B., and Mallin, M.A. 2007. Water column oxygen demand and sediment oxygen flux: patterns of oxygen depletion in tidal creeks. *Hydrobiologia* 586, 235-248.

Mallin, M.A., Esham, E.C., Williams, K.E., and Nearhoof, J.E. 1999. Tidal stage variability of fecal coliform and chlorophyll *a* concentrations in coastal creeks. *Marine Pollution Bulletin* 38, 414-422.

Mallin, M.A., Williams, K.E., Esham, E.C., and Lowe, R.P. 2000. Effect of human development on bacteriological water quality in coastal watersheds. *Ecological Applications* 10, 1047-1056.

Mallin, M.A., Parsons, D.C., Johnson, V.L., McIver, M.R., and CoVan, H.A. 2004. Nutrient limitation and algal blooms in urbanizing tidal creeks. *Journal of Experimental Marine Biology and Ecology* 298, 211-231.

Mallin, M.A., Cahoon, L.B., Toothman, B.R., Parsons, D.C., McIver, M.R., Ortwine, M.L., and Harrington, R.N. 2007. Impacts of a raw sewage spill on water and sediment quality in an urbanized estuary. *Marine Pollution Bulletin* 54, 81–88.

Mallin, M.A, McIver, M.R, Spivey, M.I.H., Tavares, M.E., Alphin, T.D., and Posey, M.H. 2008. *Environmental Quality of Wilmington and New Hanover County Watersheds, 2006-2007*. CMS Report 08-01. Center for Marine Science, UNC Wilmington.

Mallin, M.A., Bohrer, L.E., McIver, M.R., and Protopappas, S., 2013. Environmental quality of Wilmington and New Hanover County watersheds, 2012.CMS Report 13-01. Center for Marine Science, University of North Carolina Wilmington, Wilmington, NC.

Martin, M., Osborn, K.E., Billig, P., and Glickstein, N. 1981. Toxicities of ten metals to *Crassostrea gigas* and *Mytilus edulis* embryos and *Cancer magister* larvae. *Marine Pollution Bulletin* 12, 305-308.

McGee, B.L., Schlekat, C.E., Boward, D.M., and Wade, T.L., 1995. Sediment contamination and biological effects in a Chesapeake Bay marina. *Ecotoxicology* 4, 39-59.

Meador, J.P., Stein, J.E., Reichert, W.L., Varanasi, U., 1995. Bioaccumulation of polycyclic aromatic hydrocarbons by marine organisms. *Reviews of Environmental Contamination and Toxicology* 143, 79-165.

Menzie, C.A., Potocki, B.B., and Santodonato, J. 1992. Exposure to carcinogenic PAHs in the environment. *Environmental Science and Technology* 26, 1278-1284.

Mercado-Silver, N., 2005. Condition index of the eastern oyster, *Crassostrea virginica* (Gmelin, 1791) in Sapelo Island Georgia—Effects of site, position on be and pea crab parasitism. *Journal of Shellfish Research* 24,121-126.

Miller, T.R., Heidler, J., Chillrud, S.N., DeLaquil, A., Ritchie, J.C., Mihalic, J.N., Bopp, R., and Halden, R.U., 2008. Fate of triclosan and evidence for reductive dechlorination of triclocarban in estuarine sediments. *Environmental Science and Technology* 42, 4570-4576.

Morales, S., Canosa, P., Rodríguez, I., Rubí,, E., and Cela, 2005. Microwave assisted extraction followed by gas chromatography with tandem mass spectrometry for the determination of triclosan and two related chlorophenols in sludge and sediments *Journal of Chromatography A* 1082, 128–135.

North Carolina Division of Marine Fisheries, 2001. North Carolina Oyster Fishery Management Plan. North Carolina Department of Environment and Natural Resources, Morehead City, NC (available at *http://www.ncfisheries.net/download/oysFMP.pdf*).

O'Connor, T.P. 1998. Mussel Watch results from 1986 to 1996. *Marine Pollution Bulletin* 37, 14-19.

O'Connor, T.P. 2002. National distribution of chemical concentrations in mussels and oysters in the USA. *Marine Environmental Research* 53, 117-143.

Oros, D.R. and Ross, J.R.M., 2004. Polycyclic aromatic hydrocarbons in San Francisco Estuary sediments. *Marine Chemistry* 86:169-184.

Oyster Restoration and Protection Plan for North Carolina, 2007. Oyster Restoration and Protection Plan for North Carolina: A Blueprint For Action - Second Edition, 2008-2012 (available at *http://www.nccoast.org/newsroom/images-pdfs/guides/OysterPlan_08.pdf*).

Páez-Osuna, F., Frías-Espericueta, M.G., and Osuna-López, J.I., 1995. Trace metal concentrations in relation to season and gonadal maturation in the oyster *Crassostrea iridescens Marine Enviromental Research*, 40, 19-31.

Pait, A. S., De Souza, A. E., & Farrow, D. R. G. (1992). *Agricultural pesticide use in coastal areas: a national summary*. Silver Spring, MD: NOAA NOS Office of Ocean Resources Conservation and Assessment.

Presley, B.J., Taylor, R.J., and Boothe, P.N. 1990. Trace metals in Gulf of Mexico oysters. *Science of the Total Environment* 97-98, 551-593.

Rebelo, M.F.; Amaral, M.C.R., and Pfeiffer, W.C., 2005. Oyster condition index in *Crassostrea rhizophorae* (Guilding, 1828) from a heavy-metal polluted coastal lagoon. *Brazilian Journal of Biology*.65, 345-351.

Remberger, M., Sternbeck, J., and Strömberg, K., 2002. Screening av triclosan och vissa bromerade fenoliska ämnen i Sverige. IVL Rapport/report B1477, Stockholm, Sweden.

Renwick, W.H. and Ashley, G.M., 1984. Sources, storages, and sinks of fine-grained sediments in a fluvial-estuarine system. *Geological Society of America Bulletin* 95, 1343-1348.

Rice, M..A. and Rheault, R.B., Jr., 1996. Food-limited growth and condition index in the eastern oyster, Crassostrea virginica (Gmelin, 1791), and the bay scallop, Argopecten irradians irradians (Lamarck, 1819). J*ournal of Shellfish Research* 15, 271-283.

Riedel, G.F., and Valette-Silver, N., 2002. Differences in the bioaccumulation of arsenic by oysters from Southeast coastal US and Chesapeake Bay: environmental versus genetic control. *Chemosphere* 49, 27-37

Riedel, G.F., Abbe, G.R. and Sanders, J.G. 1998. Temporal and spatial variations of trace metal concentrations in oysters from the Patuxent River, Maryland. *Estuaries* 21, 423-434.

Sackett, D.K., Aday, D.D., Rice, J.A., and Cope, W.G., 2009. A statewide assessment of mercury dynamics in North Carolina water bodies and fish. *Transactions of the American Fisheries Society* 138, 1328-1341.

Sami, S., Faisal, M., and Huggett, R.J. 1992. Alterations in cytometric characteristics of hemocytes from the American oyster *Crassostrea virginica* exposed to polycyclic aromatic hydrocarbon (PAH) contaminated sediment. *Marine Biology* 113, 247-252.

Sanders, M., 1995. Distribution of Polycyclic Aromatic Hydrocarbons in Oyster (*Crassostrea virginica*) and surface sediment from two estuaries in South Carolina. *Archives of Environmental Contamination and Toxicology* 28, 397-405.

Schiff, K., Diehl, D., and Valkirs, A., 2004. Copper emissions from antifouling paint on recreational vessels. *Marine Pollution Bulletin* 48, 371-377.

Schiff, K., Brown, J., Diehl, D., and Greenstein, D., 2007. Extent and magnitude of copper contamination in marinas of the San Diego region, California, USA. *Marine Pollution Bulletin* 54, 322-328.

Shumway, S.E., 1996. Natural environmental factors, p. 467-513. *In* V. S. Kennedy, R. I. E. Newell, and A. F. Able (eds.), The Eastern Oyster, *Crassostrea virginica.* Maryland Sea Grant College,College Park, Maryland.

Suarez, S., Dodd, M.C., and Omil, F., and von Gunten, U., 2007. Kinetics of triclosan oxidation by aqueous ozone and consequent loss of antibacterial activity: Relevance to municipal wastewater ozonation.. *Water Research* 41, 2481-2490.

Sumpter, J.P. and Johnson, A.C. 2005. Lessons from endocrine disruption and their application to other issues concerning trace organics in the aquatic environment. *Environmental Science and Technology* 39, 4321-4332.

Tixier, C., Singer, H.P., Canonica, S.,a nd Müller, S.R., 2002. Phototransformation of triclosan in surface waters: A relevant elimination process for this widely ssed biocide--- Laboratory studies, field measurements, and modeling. *Environmental Science and Technology* 36, 3482-3489.

Tobiszewski, M. and Namieśnik, J., 2012. PAH diagnostic ratios for the identification of pollution emission sources. *Environmental Pollution* 162:110-119. JECFA/61/SC

United Nations Food and Agriculture Organization and World Health Organization, 2003. Joint FAO/WHO Expert Committee on Food Additives. 61st Meeting, Rome, Italy. Available at: *ftp://ftp.fao.org/es/esn/jecfa/jecfa61sc.pdf*.

United States Environmental Protection Agency, 1986. US EPA Method 8100. Polynuclear aromatic hydrocarbons. US EPA, Washington, DC.

United States Environmental Protection Agency, 1992. Methods for the determination of metals in environmental samples.Boca Raton, Florida: CRC Press, 1992.

Valette-Silver, N.J., Riedel, G.F., Crecelius, E.A., Windom, H., Smith, R.G., and Dolvin, S.S., 1999. Elevated arsenic concentrations in bivalves from the southeast coasts of the USA. *Marine Environmental Research* 48, 311-333.

Valters, K., Li, H., Alaee, M., D'Sa, I., Marsh, G., Bergman, A., and Letcher, R.J., 2005. Polybrominated diphenyl ethers and hydroxylated and methoxylated brominated and chlorinated analogues in the plasma of fish from the Detroit River. *Environmental Science and Technology* 39, 5612-5619.

Vázquez-Sauceda, M., Aguirre-Guzmán, G., Sánchez-Martínez, J.G., and Pérez-Castañeda, R., 2011. Cadmium, lead and zinc concentrations in water, sediment and oyster (*Crassostrea virginica*) of San Andres Lagoon, Mexico. *Bulletin of Environmental Contamination and Toxicology*, 86, 410–414.

Wade, T.L., Atlas, E.A., Brooks, J.M, Kennicutt, II, M.C., Fox, R.G., Sericano, J., Garcia-Romero, B., and DeFreitas, D., 1998. NOAA Gulf of Mexico Status and Trends Program: Trace organic contaminant distribution in sediments and oysters. *Estuaries* 11, 171-179.

Wauchope, R.D. and Mc Dowell, L.L., 1984. Adsorption of phosphate, arsenate, methanearsonate, and cacodylate by lake and stream sediments: Comparisons with soils. *Journal of Environmental Quality* 13, 499-504.

Weis, J.S. and Weis, P., 1992. Transfer of contaminants from CCA-treated lumber to aquatic biota. *Journal of Experimental Marine Biology and Ecology* 161, 189-199.

Weis, J.S. and Wies, P., 1993. Trophic transfer of contaminants from organisms living by chromated-copper-arsenate (CCA)-treated wood to their *predators. Journal of Experimental Marine Biology and Ecology* 168, 25-34.

Weis, J.S., Weis, P., and Proctor, T., 1998. The extent of benthic impacts of CCA-treated wood structures in Atlantic Coast estuaries. *Archives of Environmental Contamination and Toxicology* 34, 313-322.

Westerholm, R. and Li, H., 1994. A multivariate statistical analysis of fuel-related polycyclic aromatic hydrocarbon emissions from heavy-duty diesel vehicles. *Environmental Science and Technology* 28:965-972.

Wilson, B., Zhu, J., Cantwell, M., and Olsen, C.R., 2008. Short-term dynamics and retention of triclosan in the lower Hudson River Estuary. *Marine Pollution Bulletin* 56, 1230-1233.

WHO (World Health Organization), 2010. Exposure to arsenic: A major public health concern. Geneva, Switzerland. Available at: *http://www.who.int/ipcs/features/arsenic.pdf.*

Yunker, M.B., Macdonald, R.W., Vingarzan, R., Mitchell, R.H., Goyette, D., and Sylvestre, S. . 2002. PAHs in the Fraser River basin: a critical appraisal of PAH ratios as indicators of PAH source and composition. *Organic Geochemistry* 33:489-515.

Zheng, J., Hintelmann, H., Dimock, B., Dzurko, M.S., 2003. Speciation of arsenic in water, sediment, and plants of the Moira watershed, Canada, using HPLC coupled to high resolution ICP–MS. *Analytical and Bioanalytical Chemistry* 377, 14-24.

APPENDIX A

Table A1. Sample site information and measurements in oyster tissues and sediments. All trace metal concentrations are in µg g^{-1} dry weight and all organic concentrations are in ng g^{-1} dry weight. “bdl” denote below detection limit. Missing values indicate data were not collected for *that* measurement.

Site designation	Site description	Collection date (DDMMYYYY)	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
CMS	Center for Marine Science (ICW)	9/27/2011	34º 8’26"N, 77º51’48.2"W	1			8.8	22.1883	0.5920	
CMS				2			7.4	12.7329	0.2909	
CMS				3			9.3	22.9471	0.6412	
CMS				4			10.9	22.7812	0.5762	
CMS				5			6.9	8.6188	0.2717	
CMS				6			7.8	14.2175	0.3955	
CMS				7			7.3	11.0064	0.3357	
CMS				8			9.5	17.9770	0.6237	
CMS				9			8.5	13.4064	0.4271	
CMS				10			7.8	14.8522	0.4240	
CMS TM oyster composite										
CMS organic oyster composite										
CMS sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
WB-NB	Wrightsville Beach-near bridge	9/15/2011	34º13'5.5"N, 77º47'46.9"W	1	23.6391	3.9056	6.4	18.2233	0.4790	8.84
WB-NB	at Salisbury St.			2	23.8499	4.2525	7.3	17.0266	0.4914	7.20
WB-NB				3	51.4366	4.9774	7.0	30.8572	0.5774	2.81
WB-NB				4	23.7782	4.0011	7.0	15.5147	0.4321	5.23
WB-NB				5	54.7182	6.9156	10.0	40.0035	0.8343	5.67
WB-NB				6	17.0256	3.0389	6.1	11.3309	0.3663	6.43
WB-NB				7	40.7768	5.7874	7.4	27.5079	0.6874	5.18
WB-NB				8	40.4301	6.3902	8.1	28.0579	0.6559	5.30
WB-NB				9	19.7671	3.0433	6.0	14.0795	0.3247	5.71
WB-NB				10	12.7955	2.026	6.0	9.1653	0.2311	6.37
WB-NB TM oyster composite										
WB-NB sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g) g/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
CMS	1	21.6	0.99	0.23	38	0.37	6.70	0.112	1.55	0.00	1470
CMS	2	20.5	1.00	0.28	96	0.31	0.51	0.111	0.20	2.62	1810
CMS	3	23.3	0.99	0.15	83	0.11	4.05	0.092	0.83	2.80	1870
CMS	4	17.8	1.11	0.25	135	0.42	2.82	0.09	0.67	4.47	2940
CMS	5	20.3	0.96	0.21	41	0.90	0.61	0.092	0.11	0.93	1290
CMS	6	22.6	1.00	0.09	96	0.27	1.76	0.112	0.31	3.04	1920
CMS	7	19.6	0.88	0.20	108	0.14	0.78		0.34	3.79	2080
CMS	8	20.7	1.00	0.16	90	0.45	6.20		0.87	2.95	2040
CMS	9	22.7	1.07	0.21	89	0.45	2.90		0.55	2.48	1700
CMS	10	19.7	0.92	0.21	113	0.29	2.61		0.43	3.70	2600
CMS TM oyster composite		*21.0*	*1.0*	*0.20*	*89*	*0.35*	*3.47*	*0.10*	*0.69*	*2.68*	*2000*
CMS organic oyster composite											
CMS sediment sample		4.46	0.10		5.6	6.9	16.9		0.71		18.8
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
WB-NB	1	33.7	1.14	0.30	75.3	0.82	26.0	0.115		2.00	1051
WB-NB	2	33.7	1.40	0.29	143	0.63	7.90	0.088		0.64	669
WB-NB	3	24.2	1.52	0.37	115	0.25	7.34	0.112		0.74	849
WB-NB	4	28.4	1.39	0.28	204	0.11	15.9	0.086		0.85	822
WB-NB	5	34.5	1.19	0.26	135	0.06	5.86	0.095		0.19	770
WB-NB	6	33.4	1.30	0.32	196	0.63	13.3	0.124		0.40	863
WB-NB	7	34.5	1.47	0.33	172	0.45	13.1			0.04	746
WB-NB	8	36.2	0.95	0.24	236	BD	18.7			0.32	1001
WB-NB	9	40.3	1.08	0.20	81.8	0.63	5.81			0.43	1102
WB-NB	10	35.2	1.24	0.19	135	0.52	6.01			0.64	883
WB-NB TM oyster composite		*32.7*	*1.3*	*0.29*	*164*	*0.36*	*12.2*	*0.10*		*0.57*	*864*
WB-NB sediment sample											

Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
CMS									
CMS									
CMS									
CMS									
CMS									
CMS									
CMS									
CMS									
CMS									
CMS									
CMS TM oyster composite									
CMS organic oyster composite		bdl	bdl	bdl	5.9	bdl	bdl	bdl	bdl
CMS sediment sample		1.6	bdl	1.0	0.3	4.8	4.0	2.0	6.6
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
WB-NB									
WB-NB									
WB-NB									
WB-NB									
WB-NB									
WB-NB									
WB-NB									
WB-NB									
WB-NB									
WB-NB									
WB-NB TM oyster composite									
WB-NB sediment sample									

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
CMS										
CMS										
CMS										
CMS										
CMS										
CMS										
CMS										
CMS										
CMS										
CMS										
CMS TM oyster composite										
CMS organic oyster composite	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	5.9
CMS sediment sample	6.1	3.0	bdl	bdl	bdl	0.6	bdl	1.3	bdl	31.2
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
WB-NB										
WB-NB										
WB-NB										
WB-NB										
WB-NB										
WB-NB										
WB-NB										
WB-NB										
WB-NB										
WB-NB										
WB-NB TM oyster composite										
WB-NB sediment sample										

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
WC-H	Whiskey Creek head	3/23/2012	34º 9'36.8"N, 77º52'00.84"W	1			9.5	31.3825	0.3735	
WC-H				2			9.8	24.9791	0.1510	
WC-H				3			10.7	32.0016	0.3499	
WC-H				4			7.7	22.0870	0.2508	
WC-H				5			8.3	28.4143	0.4609	
WC-H				6			10.7	32.2433	0.5399	
WC-H				7			5.5	12.7504	0.2743	
WC-H				8			6.7	20.0890	0.3463	
WC-H				9			11	18.8283	0.3262	
WC-H				10			10.8	35.5143	0.6259	
WC-H TM oyster composite										
WC-H organic oyster composite										
WC-H sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
WC-M	Whiskey Creek mouth	3/6/2012	34º9'25.61"N, 77º 51'17.83"W	1			7.8	26.6593	0.58	
WC-M				2			7.7	23.5462	0.53	
WC-M				3			6.7	19.3775	0.65	
WC-M				4			7.5	24.7280	0.87	
WC-M				5			7.2	25.7585	0.59	
WC-M				6			8.9	50.5248	1.50	
WC-M				7			8.5	16.7544	1.00	
WC-M				8			9.6	28.0385	0.74	
WC-M				9			10	30.9533	0.29	
WC-M				10			10.9	48.3926	0.82	
WC-M TM oyster composite										
WC-M sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
WC-H	1	19.0	1.42		411			0.172			5050
WC-H	2	13.5	1.88		506						6310
WC-H	3	16.8	1.46		469			0.174			8940
WC-H	4	18.2	1.44		232						2730
WC-H	5	16.1	1.26		225			0.164			3340
WC-H	6	19.5	1.20		308			0.157			4240
WC-H	7	19.6	1.36		296						4720
WC-H	8	19.3	1.28		383			0.145			5100
WC-H	9	18.8	1.45		331						5090
WC-H	10	31.9	1.62		120			0.150			1730
WC-H TM oyster composite		*20.5*	*1.4*		*303*			*0.16*			*4400*
WC-H organic oyster composite											
WC-H sediment sample		7.1	0.12		11.7	8.4	30.9		0.72		34.5
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
WC-M	1	36.3	2.15		229						3070
WC-M	2	20.0	1.61		363			0.128			8640
WC-M	3	38.2	1.68		169						1930
WC-M	4	31.4	1.56		152			0.0873			2150
WC-M	5	34.4	1.77		167						2220
WC-M	6	32.3	1.67		251			0.0894			2920
WC-M	7	33.6	1.66		168						2460
WC-M	8	26.8	1.47		150						2470
WC-M	9	34.5	2.04		205			0.131			1730
WC-M	10	33.5	1.81		162						3400
WC-M TM oyster composite		*34.5*	*1.8*		*215*			*0.11*			*3230*
WC-M sediment sample		*1.336868412*	*0.013323205*		*0.636974108*	***1.541957981***	***3.541640091***		***0.052878697***		*4.36352775*

Site designation	Triclosan	napthalene	acenapthene	acenaphthylene	dibenzofuran	fluorene	anthracene	phenanthrene	fluoranthene
WC-H									
WC-H									
WC-H									
WC-H									
WC-H									
WC-H									
WC-H									
WC-H									
WC-H									
WC-H									
WC-H TM oyster composite									
WC-H organic oyster composite	206								
WC-H sediment sample	26.2								
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
WC-M									
WC-M									
WC-M									
WC-M									
WC-M									
WC-M									
WC-M									
WC-M									
WC-M									
WC-M									
WC-M TM oystser composite									
WC-M sediment sample									

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
WC-H										
WC-H										
WC-H										
WC-H										
WC-H										
WC-H										
WC-H										
WC-H										
WC-H										
WC-H										
WC-H TM oyster composite										
WC-H organic oyster composite										
WC-H sediment sample										
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
WC-M										
WC-M										
WC-M										
WC-M										
WC-M										
WC-M										
WC-M										
WC-M										
WC-M										
WC-M										
WC-M TM oystser composite										
WC-M sediment sample										

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
BC-H	Bradley Creek head	9/27/2011	34º12'29.46"N, 77º 49'22.19"W	1			7.60	24.3201	0.3769	
BC-H				2			7.00	19.1104	0.3976	
BC-H				3			8.10	43.8844*	0.2192	
BC-H				4			5.60	18.9238	0.2194	
BC-H				5			6.30	22.3079	0.5246	
BC-H				6			5.40	9.4047	0.2135	
BC-H				7			7.20	20.5731	0.2144	
BC-H				8			5.60	20.5795	0.3063	
BC-H				9			6.90	13.6929	0.2558	
BC-H				10			7.60	39.0889*	0.4903	
BC-H TM oyster composite										
BC-H organic oyster composite										
BC-H sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
BC-M	Bradley Creek mouth	3/6/2012	34º12'30.24" N, 77º 49'25.81"W	1			10.8	47.6349	1.2	
BC-M				2			8.1	50.1326	1.02	
BC-M				3			9.6	31.7383	0.98	
BC-M				4			7.7	33.4385	1.31	
BC-M				5			11.2	57.6409	0.85	
BC-M				6			9.4	48.0730	0.92	
BC-M				7			8.4	22.1894	0.84	
BC-M				8			8.9	32.1049	0.7	
BC-M				9			9.9	40.5477	1.1	
BC-M				10			10.3	108.2717	2.3	
BC-M TM oyster composite										
BC-M sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
BC-H	1	17.7	1.10		315			0.106			3680
BC-H	2	18.2	0.94		270			0.102			2310
BC-H	3	21.2	1.39		132						1340
BC-H	4	22.8	1.08		729						4690
BC-H	5	24.3	1.10		564			0.105			6200
BC-H	6	23.5	1.23		451			0.0951			3670
BC-H	7	23.3	1.28		219						1720
BC-H	8	24.5	1.21		301			0.0932			4300
BC-H	9	21.2	1.34		562						5230
BC-H	10	18.5	1.12		692			0.135			5620
BC-H TM oyster composite		*21.2*	*1.2*		*444*			*0.11*			*4180*
BC-H organic oyster composite											
BC-H sediment sample		*8.068164501*	*0.213654602*		*36.90958828*	*17.08*	*22.72*		*1.0*		*59.7617863*
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
BC-M	1	41.4	2.40		256			0.0852			3850
BC-M	2							0.0881			
BC-M	3	37.2	2.11		150			0.0769			2340
BC-M	4	33.3	1.83		126			0.0562			1940
BC-M	5	36.9	2.05		185						3090
BC-M	6	41.3	2.27		218						3490
BC-M	7	35.2	1.70		136			0.0651			1460
BC-M	8	38.7	2.22		147			0.113			2590
BC-M	9	40.7	1.91		214			0.0787			2840
BC-M	10	40.6	2.39		188			0.0823			4200
BC-M TM oyster composite		*38.6*	*2.1*		*183*			*0.080*			*3050*
BC-M sediment sample											

Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
BC-H									
BC-H									
BC-H									
BC-H									
BC-H									
BC-H									
BC-H									
BC-H									
BC-H									
BC-H									
BC-H TM oyster composite									
BC-H organic oyster composite	267	7.7	5.7	0.0	8.4	7.4	14.5	18.1	28.6
BC-H sediment sample	23.4	1.1	6.8	21.4	7.9	3.7	71.3	202.3	909.6
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
BC-M									
BC-M									
BC-M									
BC-M									
BC-M									
BC-M									
BC-M									
BC-M									
BC-M									
BC-M									
BC-M TM oyster composite									
BC-M sediment sample									

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
BC-H										
BC-H										
BC-H										
BC-H										
BC-H										
BC-H										
BC-H										
BC-H										
BC-H										
BC-H										
BC-H TM oyster composite										
BC-H organic oyster composite	28.4	28.0	25.6		5.3	49.0	31.0	9.3	11.6	278.6
BC-H sediment sample	843.6	347.4	528.7	116.7	52.9	754.5	579.0	178.0	335.6	4960.4
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
BC-M										
BC-M										
BC-M										
BC-M										
BC-M										
BC-M										
BC-M										
BC-M										
BC-M										
BC-M										
BC-M TM oyster composite										
BC-M sediment sample										

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
BH-1	Bald Head Island harbor	4/10/2012	33°52’34.89”N, 77°59’49.57”W	1		2.5548	4.25		0.3849	
BH-1				2		4.9024	6.40		0.9559	
BH-1				3		2.3718	5.75		0.4279	
BH-1				4		2.5624	5.45		0.4463	
BH-1				5		3.8285	6.10		0.7318	
BH-1				6		2.3764	6.70		0.5128	
BH-1				7		4.1666	6.40		0.7485	
BH-1				8		3.9461	6.00		0.7591	
BH-1				9		2.8517	6.10		0.5511	
BH-1 TM oyster composite										
BH-1 organic oyster composite										
BH-1 sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
BH-3	Bald Head Island upper creek	4/10/2012	33°51’31.56”N, 77°58’23.38”W	1		2.1654	5.50		0.2161	
BH-3				2		4.3869	6.15		0.5174	
BH-3				3		4.6196	6.50		0.6059	
BH-3				4		3.8045	5.90		0.4901	
BH-3				5		5.8203	7.00		0.7484	
BH-3				6		4.0948	6.30		0.4410	
BH-3				7		6.1555	6.20		0.6737	
BH-3				8		1.8965	5.75		0.2655	
BH-3				9		2.5733	5.20		0.3055	
BH-3				10		3.1758	5.00		0.3264	
BH-3 TM oyster composite										
BH-3 organic oyster composite										
BH-3 sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
BH-1	1	36.9	1.61	0.311	25.1		24.3	0.0802		0.156	459
BH-1	2	32.9	1.60	0.227	30.3		38.8	0.0816		0.354	421
BH-1	3	21.6	1.16	0.248	30.5		6.91	0.109		0.355	384
BH-1	4	23.8	1.46	0.176	29.9		6.91	0.0853		0.351	415
BH-1	5	36.3	2.02	0.308	20.5		7.47	0.088		0.149	371
BH-1	6	41.6	1.83	0.322	30.5		44.2	0.0813		0.202	291
BH-1	7	39.6	1.65	0.255	42.4		43.9	0.0834		0.583	508
BH-1	8	25.2	1.40	0.213	30.6		7.92	0.0696		0.215	606
BH-1	9	27.2	1.45	0.128	40.1		7.37	0.0749		0.257	721
BH-1 TM oyster composite		*32.1*	*1.6*	*0.242*	*31.3*		*22.4*	*0.0827*		*0.301*	*469*
BH-1 organic oyster composite											
BH-1 sediment sample											
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
BH-3	1	20.2	1.28	0.363	97.5		30.6			1.24	2616
BH-3	2	26.6	1.50	0.391	84.3		37.6	0.127		0.572	3077
BH-3	3	19.9	1.23	0.276	53.5		20.3	0.155		0.477	2182
BH-3	4	21.5	1.24	0.335	90.7		26.3	0.151		0.942	2298
BH-3	5	24.4	1.35	0.4	77.5		31	0.135		0.808	2915
BH-3	6	25.5	1.09	0.308	56.1		45.6	0.11		0.454	1403
BH-3	7	26.1	1.40	0.314	62.3		46.5	0.103		0.652	4091
BH-3	8	22.7	1.18	0.234	97.7		32			0.882	2361
BH-3	9	21.8	1.40	0.258	80.2		33.6	0.155		0.773	2411
BH-3	10	17.7	1.05	0.232	65.0		19.9	0.15		0.645	1944
BH-3 TM oyster composite		*23.1*	*1.3*	*0.321*	*73.6*		*32.9*	*0.134*		*0.706*	*2650*
BH-3 organic oyster composite											
BH-3 sediment sample											

Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
BH-1									
BH-1									
BH-1									
BH-1									
BH-1									
BH-1									
BH-1									
BH-1									
BH-1									
BH-1 TM oyster composite									
BH-1 organic oyster composite	258	bdl	bdl	bdl	12.0	bdl	bdl	bdl	bdl
BH-1 sediment sample	6.0	bdl	11.3	bdl	0.8	0.3	3.1	1.9	1.1
Site designation		**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
BH-3									
BH-3									
BH-3									
BH-3									
BH-3									
BH-3									
BH-3									
BH-3									
BH-3									
BH-3									
BH-3									
BH-3 TM oyster composite									
BH-3 organic oyster composite		bdl	bdl	bdl	0.3	bdl	bdl	bdl	bdl
BH-3 sediment sample		30.8	163.2	36.4	21.8	93.9	81.7	78.3	38.6

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
BH-1										
BH-1										
BH-1										
BH-1										
BH-1										
BH-1										
BH-1										
BH-1										
BH-1										
BH-1 TM oyster composite										
BH-1 organic oyster composite	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	12.0
BH-1 sediment sample	4.0	2.5	1.8	bdl	bdl	1.8	0.5	0.6	bdl	29.5
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
BH-3										
BH-3										
BH-3										
BH-3										
BH-3										
BH-3										
BH-3										
BH-3										
BH-3										
BH-3										
BH-3 TM oyster composite										
BH-3 organic oyster composite	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.3
BH-3 sediment sample	69.4	180.3	225.6	0.0	0.0	0.0	242.0	242.1	260.8	1765.0

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
HC-H	Hewletts Creek head	9/26/2012	34°11’32.1”N, 77°51’38.3”W	1	26.1088	5.0798	6.40	17.6295	0.3481	4.11
HC-H				2	31.4615	4.6589	6.90	22.8937	0.4479	5.23
HC-H				3	17.2039	2.4189	7.40	11.7018	0.2405	4.37
HC-H				4	22.1131	4.1856	6.70	15.2768	0.3446	5.04
HC-H				5	20.7995	4.2237	6.15	13.1505	0.3796	4.96
HC-H				6	21.1750	4.2069	6.60	14.4987	0.3360	5.03
HC-H				7	23.0138	5.3525	6.10	14.7168	0.3820	4.60
HC-H				8	18.3567	3.9598	6.25	12.0472	0.3477	5.51
HC-H				9	41.0453	7.9849	7.60	26.1016	0.7044	4.71
HC-H				10	26.6921	5.1879	7.25	18.5911	0.4332	5.35
HC-H TM oyster composite										
HC-H sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
HC-M	Hewletts Creek mouth	7/16/2012	34°10’47.7”N, 77°50’11.5”W	1	58.1137	9.0065	8.10	42.4893	0.7708	4.93
HC-M				2	75.8681	9.2111	8.95	57.6483	1.1352	6.23
HC-M				3	56.7269	8.6634	7.40	40.3163	0.9629	5.87
HC-M				4	54.2927	6.2342	6.80	42.8924	0.8544	7.49
HC-M				5	40.2317	5.7827	9.10	28.1363	0.6698	5.54
HC-M				6	55.9865	7.5901	9.20	38.4775	0.8113	4.63
HC-M				7	46.2508	5.1762	6.25	36.3476	0.6820	6.89
HC-M				8	56.2968	8.2243	9.90	40.3662	0.7243	4.55
HC-M				9	83.1004	9.3661	9.45	58.1075	1.0028	4.01
HC-M				10	56.2365	4.6232	8.20	41.8524	0.5968	4.15
HC-M TM oyster composite										
HC-M organic oyster composite										
HC-M sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
HC-H	1	18.2	1.08	0.337	97.9		7.1	0.126		0.957	3951
HC-H	2							0.13			
HC-H	3										
HC-H	4	20.4	1.08	0.309	139		12.4	0.117		0.977	4903
HC-H	5							0.112			
HC-H	6	18.9	1.05	0.284	70.8		8.84	0.122		0.256	2294
HC-H	7	21.3	0.90	0.277	64.3		11.5	0.131		0.559	1959
HC-H	8	16	0.84	0.52	98.9		11.7	0.117		0.825	7415
HC-H	9	16.3	0.97	0.447	139		11.8	0.11		1.05	7521
HC-H	10	16.3	0.93	0.421	137		18.5	0.0937		0.811	4319
HC-H TM oyster composite		*17.9*	*0.97*	*0.381*	*111*		*11.9*	*0.117*		*0.810*	*4950*
HC-H sediment sample		1.15	0.065		0.85	1.79	4.03		0.10		5.53
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
HC-M	1	41.7	2.0	0.231	22.5		8.89	0.0921		0.252	1118
HC-M	2	38	1.9	0.271	55.1		14.2	0.096		0.641	2064
HC-M	3	37.4	1.9	0.229	18.6		5.02	0.0962		0.029	1073
HC-M	4	39	1.6	0.371	42.5		9.09	0.12		0.455	2252
HC-M	5	47.3	2.1	0.268	62.9		6.85	0.107		0.639	2429
HC-M	6	41	2.0	0.235	54.6		8.35	0.105		0.447	2640
HC-M	7	42	2.1	0.237	29.2		5.37	0.099		0.242	2133
HC-M	8	35.9	2.1	0.201	7.4		8.97	0.108		0.308	500
HC-M	9	37.9	1.8	0.325	33.9		9.04	0.134		0.268	2498
HC-M	10	40.7	2.2	0.338	58.7		11.8	0.106		0.495	1976
HC-M TM oyster composite		*39.8*	*2.0*	*0.271*	*38.4*		*8.91*	*0.107*		*0.375*	*1880*
HC-M organic oyster composite											
HC-M sediment sample		3.03	0.073		3.78	5.72	13.5		0.50		14.9

Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
HC-H									
HC-H									
HC-H									
HC-H									
HC-H									
HC-H									
HC-H									
HC-H									
HC-H									
HC-H									
HC-H TM oyster composite									
HC-H sediment sample									
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
HC-M									
HC-M									
HC-M									
HC-M									
HC-M									
HC-M									
HC-M									
HC-M									
HC-M									
HC-M									
HC-M TM oyster composite									
HC-M organic oyster composite	69.4	8.6	bdl	bdl	33.6	bdl	bdl	bdl	bdl
HC-M sediment sample	36.1	10.0	bdl	bdl	bdl	bdl	1.2	bdl	26.5

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
HC-H										
HC-H										
HC-H										
HC-H										
HC-H										
HC-H										
HC-H										
HC-H										
HC-H										
HC-H										
HC-H TM oyster composite										
HC-H sediment sample										
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
HC-M										
HC-M										
HC-M										
HC-M										
HC-M										
HC-M										
HC-M										
HC-M										
HC-M										
HC-M										
HC-M TM oyster composite										
HC-M organic oyster composite	bdl	bdl	bdl	bdl	1.9	2.6	bdl	bdl	bdl	46.7
HC-M sediment sample	63.4	bdl	13.6	bdl	5.0	17.6	bdl	2.4	bdl	139.6

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
HC-N	Just north of Hewletts Creek in ICW	7/13/2012	34°11’44.0”N, 77°49’39.6”W	1	26.4928	2.9497	7.40	20.8722	0.3795	6.75
HC-N				2	30.9066	4.9867	8.15	22.5211	0.5216	6.22
HC-N				3	13.3721	1.9774	5.90	10.6311	0.2439	8.90
HC-N				4	31.6418	3.7568	7.40	22.2262	0.3745	3.98
HC-N				5	38.3532	7.2257	10.55	27.7391	0.8244	7.77
HC-N				6	21.0772	3.3629	6.25	15.1353	0.3375	5.68
HC-N				7	28.0373	3.6156	5.80	22.7923	0.3811	7.27
HC-N				8	24.9000	3.7481	8.45	19.0486	0.4458	7.62
HC-N				9	40.5918	4.7357	8.85	32.4796	0.5908	7.28
HC-N				10	36.8641	5.7767	9.25	27.6688	0.6966	7.58
HC-N TM oyster composite										
HC-N sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
PC-H	Pages Creek head	7/16/2012	34°16’39.2”N, 77°46’41.5"W	1	30.3573	5.3695	7.15	21.0173	0.4941	5.29
PC-H				2	64.0561	8.1155	8.50	47.5970	0.5832	3.54
PC-H				3	30.7840	5.6080	7.45	21.6084	0.4789	5.22
PC-H				4	25.5003	3.9983	6.50	19.3442	0.3914	6.36
PC-H				5	34.6765	5.9389	6.75	24.5899	0.5344	5.30
PC-H				6	44.6367	8.2108	8.75	31.1140	0.7122	5.27
PC-H				7	52.1292	8.1014	9.20	37.8752	0.6558	4.60
PC-H				8	18.6124	3.7419	5.90	13.5698	0.3791	7.52
PC-H				9	20.3145	3.0643	7.15	13.1942	0.3673	5.16
PC-H				10	48.6530	3.2732	7.10	36.4109	0.4359	3.56
PC-H TM oyster composite										
PC-H organic oyster composite										
PC-H sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
HC-N	1	36.5	1.9	0.192	67		7.05	0.128		1.02	2666
HC-N	2	41.9	1.8	0.196	142		9.58	0.103		1.35	2167
HC-N	3	44	1.8	0.199	89.2		4.52			1.02	1324
HC-N	4	37	1.9	0.118	81.7		4.46	0.127		0.405	1824
HC-N	5	46.8	2.0	0.18	85.3		7.24	0.0994		0.994	1806
HC-N	6	41.8	1.9	0.174	92.1		5.87	0.1		0.839	1699
HC-N	7	39.6	1.5	0.189	27.1		2.92	0.138		0	596
HC-N	8	53.4	2.9	0.148	142		5.28	0.112		1.28	2891
HC-N	9	53.9	2.3	0.299	94.5		8.67	0.106		0.735	2461
HC-N	10	44.9	2.4	0.251	83.9		20.8	0.103		1.08	2182
HC-N TM oyster composite		*44.8*	*2.1*	*0.201*	*92*		*8.65*	*0.11*		*0.907*	*2020*
HC-N sediment sample											
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
PC-H	1	28.3	1.1	0.22	87.7		9.11	0.118		1.2	1394
PC-H	2	42.3	1.9	0.329	60		4.08	0.181		3.23	2069
PC-H	3	32.7	1.3	0.15	78.7		7.68	0.100		0.615	1569
PC-H	4	27.5	1.0	0.14	95.1		8.08	0.0975		0.44	1187
PC-H	5	31.5	1.5	0.19	90.4		10.3	0.121		0.925	2515
PC-H	6	28.9	2.6	0.181	80.6		8.66	0.114		0.76	2261
PC-H	7	33	1.5	0.251	75		7.52	0.151		1.92	2381
PC-H	8	34.9	1.2	0.215	85.8		6.23	0.13		1.34	1913
PC-H	9	36.8	1.3	0.299	60.1		8.5	0.109		0.405	1364
PC-H	10	30.9	1.4	0.306	114		10.3	0.160		0.982	3450
PC-H TM oyster composite		*32.6*	*1.6*	*0.228*	*82*		*8.01*	*0.130*		*1.26*	*2060*
PC-H organic oyster composite											
PC-H sediment sample		2.89	0.084		3.40	4.50	10.4		0.59		14.9

Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
HC-N									
HC-N									
HC-N									
HC-N									
HC-N									
HC-N									
HC-N									
HC-N									
HC-N									
HC-N									
HC-N TM oyster composite									
HC-N sediment sample									
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
PC-H									
PC-H									
PC-H									
PC-H									
PC-H									
PC-H									
PC-H									
PC-H									
PC-H									
PC-H									
PC-H									
PC-H TM oyster composite									
PC-H organic oyster composite		3.5	1.4	4.6	2.3	30.0	6.8	13.2	11.1
PC-H sediment sample		#VALUE!	0.0008	0.0000	0.0000	0.0519	0.0021	0.0013	0.0005

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
HC-N										
HC-N										
HC-N										
HC-N										
HC-N										
HC-N										
HC-N										
HC-N										
HC-N										
HC-N										
HC-N TM oyster composite										
HC-N sediment sample										
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
PC-H										
PC-H										
PC-H										
PC-H										
PC-H										
PC-H										
PC-H										
PC-H										
PC-H										
PC-H										
PC-H TM oyster composite										
PC-H organic oyster composite	11.1	1.6	2.6	bdl	bdl	bdl	bdl	bdl	bdl	75.0
PC-H sediment sample	0.0045	0.0020	0.0011	0.0012	0.0000	0.0000	0.0000	0.0014	0.0007	0.1

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
PC-M	Pages Creek mouth	7/16/2012	34°16’10.6”N, 77°46’06.9”W	1	53.7615	4.9086	8.80	40.4506	0.6187	4.65
PC-M				2	21.2570	4.5300	5.90	15.6243	0.4385	7.78
PC-M				3	23.1241	2.9392	7.50	15.3011	0.3675	4.70
PC-M				4	21.4095	5.1465	6.80	14.5413	0.5100	7.43
PC-M				5	58.8064	6.1371	10.10	40.2915	0.9040	4.88
PC-M				6	35.3665	6.9672	8.40	25.3066	0.6628	6.59
PC-M				7	36.7213	4.9639	7.10	27.0429	0.5501	5.68
PC-M				8	34.1500	3.4510	8.00	24.6800	0.4901	5.18
PC-M				9	22.7813	2.8409	6.80	16.0403	0.3488	5.17
PC-M				10	85.1901	6.6724	10.20	59.8383	0.8469	3.34
PC-M TM oyster composite										
PC-M oyster composite										
PC-M sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
HoC-H	Howe Creek head	9/26/2012	34°15’17.5”N, 77°47’47.8”W	1	42.1700	9.6478	8.25	27.9352	0.7310	5.14
HoC-H				2	39.9787	9.5918	8.90	26.3664	0.8098	5.95
HoC-H				3	39.7923	9.2330	8.10	27.5247	0.7942	6.47
HoC-H				4	34.2210	6.7053	7.00	24.0756	0.6437	6.34
HoC-H				5	42.2751	7.6978	9.05	26.5709	0.6781	4.32
HoC-H				6	39.7296	9.2854	8.70	26.6775	0.8505	6.52
HoC-H				7	55.1980	13.2567	9.25	36.6565	1.2901	6.96
HoC-H				8	35.9872	9.3277	9.30	21.5082	0.8783	6.07
HoC-H				9	28.6400	5.1712	7.55	19.3783	0.5492	5.93
HoC-H				10	21.6280	3.7829	6.10	15.2147	0.4165	6.49
HoC-H TM oyster composite										
HoC-H sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
PC-M	1	29.7	1.3	0.171	85.9		11.4	0.16		0.668	2266
PC-M	2	33.3	1.4	0.211	86		6.69	0.103		0.819	2400
PC-M	3	34.1	1.5	0.175	98.5		10.3	0.125		0.646	2237
PC-M	4	28.2	1.3	0.204	63.1		6.35	0.112		0.241	1461
PC-M	5	36.5	1.6	0.241	77.1		8.43	0.058		0.443	2208
PC-M	6	24.8	1.0	0.185	50.3		10.2	0.105		0.511	1222
PC-M	7	38.1	1.4	0.199	53.8		4.77	0.142		0.773	1331
PC-M	8	37.7	1.6	0.233	83.7		4.7	0.133		0.897	2417
PC-M	9	36.8	1.6	0.191	107		8.88	0.14		1.13	2187
PC-M	10	35.1	1.6	0.314	61.9		6.47	0.192		0.895	1781
PC-M TM oyster composite		*33.3*	*1.4*	*0.22*	*73.6*		*7.83*	*0.126*		*0.678*	*1920*
PC-M oyster composite											
PC-M sediment sample		2.92	0.10		3.70	5.22	15.1		0.67		15.0
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
HoC-H	1	27.2	1.4	0.323	92.4		23.7	0.1		0.631	3214
HoC-H	2	32.6	1.3	0.25	86.9		18.6	0.118		0.525	2794
HoC-H	3	29.1	1.2	0.282	107		12.2	0.0866		0.76	2419
HoC-H	4	32	1.2	0.241	57.9		9.9	0.102		0.257	2003
HoC-H	5	29.1	1.2	0.446	149		12.5	0.131		1.06	3451
HoC-H	6	28.7	1.2	0.252	103		13.5	0.114		0.712	3421
HoC-H	7	37	1.4	0.261	65.5		13.3	0.106		0.237	2155
HoC-H	8	26	1.1	0.213	115		16.8	0.0936		0.767	2160
HoC-H	9	35.2	1.4	0.188	98.3		7.05	0.11		0.632	1938
HoC-H	10	37.5	1.4	0.158	55.1		3.84	0.104		0.209	2189
HoC-H TM oyster composite		*31.4*	*1.3*	*0.265*	*93.1*		*13.8*	*0.106*		*0.575*	*2580*
HoC-H sediment sample											

Site designation	Triclosan	napthalene	acenapthene	acenaphthylene	dibenzofuran	fluorene	anthracene	phenanthrene	fluoranthene
PC-M									
PC-M									
PC-M									
PC-M									
PC-M									
PC-M									
PC-M									
PC-M									
PC-M									
PC-M									
PC-M TM oyster composite									
PC-M organic oyster composite		7.4	1.1	5.2	0.2	28.5	5.5	6.4	3.4
PC-M sediment sample		0.0	0.0	bdl	0.3	0.3	4.4	2.0	12.2
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
HoC-H									
HoC-H									
HoC-H									
HoC-H									
HoC-H									
HoC-H									
HoC-H									
HoC-H									
HoC-H									
HoC-H									
HoC-H TM oyster composite		bdl	bdl	bdl	22.0	bdl	bdl	bdl	bdl
HoC-H sediment sample		5.8	bdl	bdl	bdl	bdl	bdl	4.1	bdl

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
PC-M										
PC-M										
PC-M										
PC-M										
PC-M										
PC-M										
PC-M										
PC-M										
PC-M										
PC-M										
PC-M TM oyster composite										
PC-M organic oyster composite	7.0	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	64.8
PC-M sediment sample	9.5	bdl	25.1	bdl	2.1	6.5	4.4	3.0	1.5	71.4
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
HoC-H										
HoC-H										
HoC-H										
HoC-H										
HoC-H										
HoC-H										
HoC-H										
HoC-H										
HoC-H										
HoC-H										
HoC-H TM oyster composite	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	22.0
HoC-H sediment sample	4.7	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	14.5

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
HoC-M	Howe Creek mouth	7/16/2012	34°14’37.0”N, 77°47’22.3”W	1	32.7756	4.5627	6.50	24.6102	0.5119	6.27
HoC-M				2	40.4445	4.8415	7.45	30.4676	0.6202	6.22
HoC-M				3	32.4960	3.4642	7.70	23.6276	0.4786	5.40
HoC-M				4	61.9419	8.1838	9.90	41.4093	0.7285	3.55
HoC-M				5	35.7995	2.8434	7.45	28.1512	0.4477	5.85
HoC-M				6	27.0180	2.8484	6.65	19.9230	0.3742	5.27
HoC-M				7	39.5243	7.4007	8.65	25.9075	0.6558	4.82
HoC-M				8	25.2525	3.0199	7.90	16.8016	0.3644	4.31
HoC-M				9	41.0721	2.8905	9.50	29.1111	0.4213	3.52
HoC-M				10	37.1592	6.8717	7.10	27.6997	0.5037	5.32
HoC-M TM oyster composite										
HoC-M sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
LWF-1	Lockwood Folly estuary site 1	9/12/2012	33°55'34.3"N, 78°13'12.5"W	1	45.5451	4.8219	74	31.4505	0.4928	3.50
LWF-1				2	34.516	3.1699	60	24.7416	0.3272	3.35
LWF-1				3	25.0752	1.9697	62	18.8940	0.2372	3.84
LWF-1				4	59.6677	6.1652	86	42.4434	0.626	3.63
LWF-1				5	39.745	1.5434	70	26.8888	0.1805	1.40
LWF-1				6	65.843	5.7977	94	47.8025	0.4911	2.72
LWF-1				7	46.2125	4.9181	73	31.6329	0.5214	3.58
LWF-1				8	34.3155	2.8726	73	23.3562	0.2136	1.95
LWF-1				9	42.2115	4.0992	66	28.7609	0.4428	3.29
LWF-1				10	47.3532	5.9001	67	31.0826	0.5505	3.38
LWF-1 TM oyster composite										
LWF-1 organic oyster composite										
LWF-1 sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
HoC-M	1	28.4	1.1	0.196	103		5.89	0.13		0.456	2061
HoC-M	2	27.9	1.1	0.161	94.8		6.93	0.104		0.526	2227
HoC-M	3	30.1	1.4	0.212	150		5.83	0.121		0.52	2483
HoC-M	4	23.6	1.2	0.176	182		8.75	0.136		1.57	3074
HoC-M	5	30.9	1.2	0.19	106		3.76	0.132		0.583	1938
HoC-M	6	29.5	1.2	0.224	133		13	0.119		0.712	2460
HoC-M	7	26.0	1.1	0.186	110		9.54	0.104		0.713	2001
HoC-M	8	31.8	1.4	0.254	172		6.77	0.113		1.26	2657
HoC-M	9	27.6	1.4	0.222	171		14.1	0.123		1.03	2658
HoC-M	10	25.0	1.1	0.142	65.2		7.37	0.131		0.297	1546
HoC-M TM oyster composite		*27.6*	*1.2*	*0.192*	*128*		*8.1*	*0.121*		*0.781*	*2320*
HoC-M sediment sample											
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
LWF-1	1	30.3	1.5		63.5		10.2	0.080	0.56		1441
LWF-1	2	33.5	2.0		46.0		5.3		0.38		1185
LWF-1	3	34.7	2.1		40.0		12.0		0.04		984
LWF-1	4	33.8	2.1		33.0		6.8	0.077	1.84		1152
LWF-1	5	23.2	1.6		47.0		13.9		0.08		1395
LWF-1	6				13.4			0.122	BD		330
LWF-1	7	24.9	1.5		41.3		12.1	0.081	0.10		1005
LWF-1	8	32.8	2.1		15.2		6.9		0.57		379
LWF-1	9	35.8	2.0		88.3		12.6	0.104	0.51		1863
LWF-1	10	31.2	2.3		27.5		6.8	0.079	0.51		1052
LWF-1 TM oyster composite		*28.1*	*1.9*		*41.7*		*8.6*	*0.090*	*0.47*		*1078*
LWF-1 organic oyster composite											
LWF-1 sediment sample											

Site designation	Triclosan	napthalene	acenapthene	acenaphthylene	dibenzofuran	fluorene	anthracene	phenanthrene	fluoranthene
HoC-M									
HoC-M									
HoC-M									
HoC-M									
HoC-M									
HoC-M									
HoC-M									
HoC-M									
HoC-M									
HoC-M									
HoC-M TM oyster composite									
HoC-M sediment sample									
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1									
LWF-1 TM oyster composite									
LWF-1 organic oyster composite		1.4	0.0	0.0	1.3	0.0	0.0	0.3	0.0
LWF-1 sediment sample		0.7	1.4	1.5	1.5	1.2	2.5	3.2	10.6

Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
HoC-M										
HoC-M										
HoC-M										
HoC-M										
HoC-M										
HoC-M										
HoC-M										
HoC-M										
HoC-M										
HoC-M										
HoC-M TM oyster composite										
HoC-M sediment sample										
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
LWF-1										
LWF-1										
LWF-1										
LWF-1										
LWF-1										
LWF-1										
LWF-1										
LWF-1										
LWF-1										
LWF-1										
LWF-1 TM oyster composite										
LWF-1 organic oyster composite	1.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	4.6
LWF-1 sediment sample	9.5	3.2	5.6	0.5	0.1	7.4	4.5	2.4	0.3	56.1

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
LWF-2	Lockwood Folly estuary site 2	9/12/2012	33°56'33.0"N, 78°13'8.4"W	1	42.0783	2.808	90	29.9110	0.3392	2.79
LWF-2				2	40.809	2.3227	79	28.8557	0.3529	2.95
LWF-2				3	34.7715	3.0353	78	24.5520	0.3808	3.73
LWF-2				4	26.4825	2.4486	65	19.5298	0.3225	4.64
LWF-2				5	31.6061	3.7405	66	22.9081	0.4213	4.84
LWF-2				6	39.5672	2.897	75	28.1747	0.3548	3.11
LWF-2				7	51.0026	3.3675	86	37.1566	0.4111	2.97
LWF-2				8	11.0217	1.2344	47	8.5067	0.1962	7.80
LWF-2				9	26.0849	1.6897	60	20.4409	0.2048	3.63
LWF-2				10	41.5814	2.4566	79	29.1524	0.2592	2.09
LWF-2 TM oyster composite										
LWF-2 organic oyster composite										
LWF-2 sediment sample										
Site designation	**Site description**	**Collection date**	**Coordinates**	**Sample**	**Whole oyster wt. (g)**	**Wet tissue wt. (g)**	**Shell length (cm)**	**Dry shell wt. (g)**	**Dry tissue wt. (g)**	**Condition index**
LWF-3	Lockwood Folly estuary site 3	9/12/2012	33°56'39.9"N, 78°13'19.9"W	1	31.3619	5.9302	93		0.4910	
LWF-3				2	27.5643	4.0553	81		0.4406	
LWF-3				3	43.3578	4.3233	95		0.4979	
LWF-3				4	42.6154	4.8691	95		0.5960	
LWF-3				5	23.3123	2.3093	75		0.3623	
LWF-3				6	36.341	4.1493	88		0.4579	
LWF-3				7	43.5846	6.5626	95		0.7280	
LWF-3				8	56.1743	4.0556	100		0.4953	
LWF-3				9	39.935	4.3212	75		0.4338	
LWF-3				10	30.8271	2.7681	81		0.3606	
LWF-3 organic oyster composite										
LWF-3 organic oyster composite										
LWF-3 sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
LWF-2	1	26.5	2.0		64.5	0.16	11.2	0.113	1.06		2468
LWF-2	2	22.7	2.0		46.2	0.89	18.8	0.112	1.62		2443
LWF-2	3	25.9	1.7		84.5	0.89	11.9	0.098	1.02		2552
LWF-2	4	32.6	1.9		60.6	0.45	8.7		0.85		1836
LWF-2	5	32.0	1.7		42.7	1.53	23.0		1.00		1537
LWF-2	6	26.6	2.2		59.6	0.01	5.6	0.098	0.62		1915
LWF-2	7	20.6	1.5		39.3	BD	11.5	0.109	0.79		1278
LWF-2	8	26.5	1.7		82.0	0.99	8.4		1.17		1833
LWF-2	9	28.3	1.2		64.0	1.05	17.2		0.91		1589
LWF-2	10	21.2	1.8		90.6	BD	4.7		0.10		2472
LWF-2 TM oyster composite		*26.2*	*1.8*		*62.5*	*0.56*	*12.1*	*0.11*	*0.90*		*2019*
LWF-2 organic oyster composite											
LWF-2 sediment sample		2.32	0.022		0.98	3.28	13.4		0.23		7.28
Site designation	**Sample**	**Arsenic (µg/g)**	**Cadmium (µg/g)**	**Cobalt (µg/g)**	**Copper (µg/g)**	**Lead (µg/g)**	**Manganese (µg/g)**	**Mercury (µg/g)**	**Nickel (µg/g)**	**Silver (µg/g)**	**Zinc (µg/g)**
LWF-3	1	34.9	2.1		187		14.33		0.54		2778
LWF-3	2	30.0	1.3		129		9.21		0.27		1949
LWF-3	3	35.2	2.1		92.5		18.28		0.40		1610
LWF-3	4	25.3	1.7		168		9.74		0.31		3356
LWF-3	5				23.9						905
LWF-3	6	24.6	1.6		155	BD	22.02		0.64		2688
LWF-3	7	29.7	1.7		114	0.94	17.68		1.26		2767
LWF-3	8	23.6	1.7		123	0.13	8.68		0.62		2299
LWF-3	9	24.5	1.8		165	0.97	11.91		1.47		3372
LWF-3	10	22.3	1.6		184	BD	17.12		1.17		3007
LWF-3 TM oyster composite		*27.8*	*1.7*		*134*	*0.42*	*14.4*		*0.75*		*2473*
LWF-3 organic oyster composite											
LWF-3 sediment sample		3.71	0.049		3.71	4.58	35.5		0.51		15.4

Site designation	Triclosan	napthalene	acenapthene	acenaphthylene	dibenzofuran	fluorene	anthracene	phenanthrene	fluoranthene
LWF-2									
LWF-2									
LWF-2									
LWF-2									
LWF-2									
LWF-2									
LWF-2									
LWF-2									
LWF-2									
LWF-2									
LWF-2 TM oyster composite									
LWF-2 organic oyster composite		6.9	0.0	0.0	3.3	1.6	0.0	3.7	0.0
LWF-2 sediment sample		0.2	0.3	0.2	0.6	0.0	1.6	3.8	6.8
Site designation	**Triclosan**	**napthalene**	**acenapthene**	**acenaphthylene**	**dibenzofuran**	**fluorene**	**anthracene**	**phenanthrene**	**fluoranthene**
LWF-3									
LWF-3									
LWF-3									
LWF-3									
LWF-3									
LWF-3									
LWF-3									
LWF-3									
LWF-3									
LWF-3									
LWF-3 TM oyster composite									
LWF-3 organic oyster composite		12.6	0.0	14.3	18.4	0.0	25.7	42.7	20.7
LWF-3 sediment sample		1.7	0.0	1.1	2.2	0.0	9.2	9.5	28.1

Site designation	pyrene	benzo(a)anthracene	chrysene	indeno(1,2,3-cd)pyrene	dibenzo(a,h)anthracene	benzo(b)fluoranthene	benzo(k)fluoranthene	benzo(a)pyrene	benzo(g.h.i)perylene	Sum of PAHs (ng/g dry weight):
LWF-2										
LWF-2										
LWF-2										
LWF-2										
LWF-2										
LWF-2										
LWF-2										
LWF-2										
LWF-2										
LWF-2										
LWF-2 TM oyster composite										
LWF-2 organic oyster composite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	15.5
LWF-2 sediment sample	7.6	3.1	3.2	0.9	0.0	6.7	2.2	1.8	0.0	39.0
Site designation	**pyrene**	**benzo(a)anthracene**	**chrysene**	**indeno(1,2,3-cd)pyrene**	**dibenzo(a,h)anthracene**	**benzo(b)fluoranthene**	**benzo(k)fluoranthene**	**benzo(a)pyrene**	**benzo(g.h.i)perylene**	**Sum of PAHs (ng/g dry weight):**
LWF-3										
LWF-3										
LWF-3										
LWF-3										
LWF-3										
LWF-3										
LWF-3										
LWF-3										
LWF-3										
LWF-3										
LWF-3 TM oyster composite										
LWF-3 organic oyster compo	0.0	40.3	28.6	11.2	16.2	34.4	38.0	19.2	14.9	337.1
LWF-3 sediment sample	21.6	14.5	13.8	23.1	1.0	15.5	11.7	11.0	7.0	170.8

Site designation	Site description	Collection date	Coordinates	Sample	Whole oyster wt. (g)	Wet tissue wt. (g)	Shell length (cm)	Dry shell wt. (g)	Dry tissue wt. (g)	Condition index
LWF-4	Lockwood Folly estuary site 4	9/12/2012	33°55'49.4"N, 78°12'58.8"W	1	37.0273	2.5439	72	20.4511	0.3423	2.07
LWF-4				2	46.3847	3.1922	79	19.1705	0.3713	1.36
LWF-4				3	59.7355	4.8665	90	29.5619	0.4806	1.59
LWF-4				4	55.2874	2.791	85	27.0900	0.2955	1.05
LWF-4				5	51.101	5.2205	85	16.2416	0.6357	1.82
LWF-4				6	43.5113	3.9082	70	24.9774	0.4845	2.61
LWF-4				7	43.6006	5.5006	85	30.3156	0.5632	4.24
LWF-4				8	34.9936	4.0263	80		0.3678	
LWF-4				9	25.2788	1.8699	70		0.2724	
LWF-4				10	41.7688	2.4608	73	22.3266	0.3089	1.59
LWF-4 TM oyster composite										
LWF-4 organic oyster composite										
LWF-4 sediment sample										

Site designation	Sample	Arsenic (µg/g)	Cadmium (µg/g)	Cobalt (µg/g)	Copper (µg/g)	Lead (µg/g)	Manganese (µg/g)	Mercury (µg/g)	Nickel (µg/g)	Silver (µg/g)	Zinc (µg/g)
LWF-4	1	17.1	1.8		23.1	0.04	9.38	0.156	0.69		829
LWF-4	2	28.0	2.1		25.9	0.52	11.34		1.44		797
LWF-4	3	31.4	2.4		47.3	0.35	6.70	0.182	0.83		1371
LWF-4	4	26.7	2.2		33.5	BD	7.61	0.154	1.13		1315
LWF-4	5	35.3	1.8		23.9	BD	12.32	0.083	0.75		879
LWF-4	6	29.0	1.9		59.3	BD	8.90	0.097	0.66		973
LWF-4	7	30.2	1.8		17.7	BD	7.48	0.089	0.74		532
LWF-4	8	28.1	1.7		22.7	BD	6.17	0.093	0.40		520
LWF-4	9	28.1	1.6		18.7	BD	15.57	0.077	0.80		469
LWF-4	10	21.2	2.0		19.5	BD	15.74	0.138	0.36		
LWF-4 TM oyster composite		*27.8*	*1.9*		*29.0*	*0.09*	*10.2*	*0.119*	*0.78*		*855*
LWF-4 organic oyster composite											
LWF-4 sediment sample		4.1	0.04		4.4	5.1	38.7		0.54		17.8

Site designation	Triclosan	napthalene	acenapthene	acenaphthylene	dibenzofuran	fluorene	anthracene	phenanthrene	fluoranthene
LWF-4									
LWF-4									
LWF-4									
LWF-4									
LWF-4									
LWF-4									
LWF-4									
LWF-4									
LWF-4									
LWF-4									
LWF-4 TM oyster composite									
LWF-4 organic oyster composite		10.4	0.0	1.8	16.8	5.0	28.7	0.0	0.0
LWF-4 sediment sample		0.1	0.1	0.3	0.5	0.2	1.3	1.8	4.3

Site designation	pyrene	benzo(a)anthracene	chrysene	indeno(1,2,3-cd)pyrene	dibenzo(a,h)anthracene	benzo(b)fluoranthene	benzo(k)fluoranthene	benzo(a)pyrene	benzo(g.h.i)perylene	Sum of PAHs (ng/g dry weight):
LWF-4										
LWF-4										
LWF-4										
LWF-4										
LWF-4										
LWF-4										
LWF-4										
LWF-4										
LWF-4										
LWF-4										
LWF-4 TM oyster composite										
LWF-4 organic oyster compo	0.0	15.2	0.0	79.2	63.6	3.7	6.8	42.0	19.7	292.9
LWF-4 sediment sample	3.6	1.7	2.9	1.8	0.2	3.3	4.0	1.4	1.3	28.9