



An Assessment of Southeast United States Headwater Tidal Creek Sediment Contamination Over a Twenty-Year Period in Relation to Coastal Development

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Abstract

Headwater tidal creeks are a primary link between estuarine and upland habitats, serving as conduits for runoff. They are sentinel habitats, providing early warning of potential harm, thus ideal systems to evaluate the effects of coastal suburban and urban development on environmental quality. Estuarine sediments have concentrations of metals, polycyclic aromatic hydrocarbons (PAHs), pesticides, polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) that are associated with human activity. High concentrations of contaminants can impair faunal communities, habitat quality, and ecosystem function. Forty-three headwater creeks were sampled between 1994 and 2006 to assess contaminants, and 18 of these were sampled again in 2014/2015. Watersheds were classified as forested, forested to suburban, suburban, or urban land. These values are based on their percent impervious cover (IC) levels and change in IC from 1994–2014. Analyses of temporal data resulted in significant relationships between IC and select metals, PAHs, pesticides, PCBs, and PBDEs. In addition, 11 of the creeks sampled in 2014/2015 have paired data from 1994/1995, allowing for change analysis over the 20 years. Results indicated increasing chemical contamination occurring with increasing levels of development, although only PAHs and total dichloro-diphenyl-trichloroethane (DDT) exhibited a statistically significant increase over time; PAHs also exhibited significantly higher concentrations in developed creeks. Additionally, several metals were deemed enriched in developed creeks based on reference conditions. These results expand our knowledge of how these systems respond to urban development and can inform managers about how human population growth along coastlines may predict altered tidal creek health.

Keywords Estuary · Atlantic · Organic compounds · Inorganic compounds · Stormwater

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Introduction

The salt marsh ecosystem is one of the most ecologically diverse and important ecosystems along the southeast United States coastline (Wiegert and Freeman 1990). The southeastern states of North Carolina, South Carolina, and Georgia support nearly two-thirds of salt marsh habitat along the east coast, with South Carolina and Georgia each having around 154,000 hectares of salt marshes and tidal creeks, and North Carolina having around 120,000 hectares (Tiner 2013). A key component of the salt marsh ecosystem are the tidal creeks which cut through and drain the marsh, and help support a diverse array of aquatic, terrestrial, and avian wildlife. In the Southeast, these creeks can range from <1 km to >10 km in length and rarely exceed three meters of depth at high tide (Mallin and Lewitus 2004). They are particularly important as primary nursery habitat for a

number of recreationally and commercially important fish and shellfish (Wiegert and Freeman 1990). Tidal creeks provide juvenile organisms with shallow protected habitat which allows them to escape predators, feed in relative safety, and seek refuge while maturing.

Metals, polycyclic aromatic hydrocarbons (PAHs), pesticides, polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) are widely distributed in the salt marsh and tidal creek environment (Vernberg et al. 1992; Sanders 1995; Kucklick et al. 1997; Sanger et al. 1999a; Sanger et al. 2015; Artigas et al. 2017). Trace metals (e.g., copper and zinc) and a few PAHs (e.g., retene) naturally occur at levels that are non-toxic to flora and fauna (Taylor 1974; Abdel-Shafy and Mansour 2016; Rigaud et al. 2019) and support the growth and survival of marine invertebrates. However, these naturally occurring compounds, as well as other PAHs, pesticides, PCBs, and PBDEs, can be enriched via anthropogenic means, such as industrial activities, agricultural runoff, vehicular emissions, and the combustion of wood (Grabe and Barron 2004; Zhang et al. 2013; Abdel-Shafy and Mansour 2016; Malik and Ravindran 2020). PAHs are known to have lethal, carcinogenic, and mutagenic properties, and benthic invertebrates can readily obtain them through the fine-grained sediment they consume during feeding (Eisler 1987; Mayer-Pinto et al. 2020). PCBs, PBDEs, and historical pesticides, such as DDT, negatively impact species survival and diversity, disrupt reproduction, and alter behavioral and physiological activities (Simpson and Roger 1995; Prince et al. 2021).

The primary objectives of this study were to assess the current relationships between southeastern headwater tidal creek sediment contamination and the level of surrounding watershed development and to compare those relationships to historical data from the 20-year Tidal Creek Project (TCP) developed by Holland et al. (2004) and refined by Sanger et al. (2015). The TCP established a conceptual model outlining the environmental responses of southeastern tidal creeks to increases in coastal development (e.g., human population density, watershed impervious cover, and stormwater runoff). Holland et al. (2004) and Sanger et al. (2015) found that when the percentage of impervious cover (IC) within a defined watershed (used as the indicator of coastal development) reaches 10–20%, physical and chemical environmental factors may be impacted, including measurable increases in sediment contamination of PAHs, trace metals, and PBDEs. When IC reaches 20–30% in a watershed, changes were observed in the living resources, particularly in the macrobenthic community with regard to community composition.

Over the 20-year lifespan of the TCP (1994–2015), several of the tidal creek watersheds sampled have experienced significant increases in watershed scale properties,

such as the amount of IC and the number of stormwater ponds. During this period of significant development, the use of best management practices (BMPs) was more extensive (e.g., implementing buffer zones and retention ponds), particularly surrounding the creeks which experienced a change of watershed classification from forested to suburban. These BMPs are meant to slow the rate of stormwater runoff and aid in the settling of particulates and contaminants before they reach nearby waterways (Muthukrishnan et al. 2005). The increased use of BMPs is expected to mitigate some of the effects of new development. Thus, this study aimed to provide information to improve our current understanding of the relationships between watershed development, the implementation of BMPs, and the changes in sediment quality. To accomplish these objectives, sediment contaminant and composition data from 18 headwater tidal creeks were collected in 2014 and 2015 in the Southeast US. These data were compared to historical TCP data.

Materials and Methods

Study Area

Headwater tidal creeks, or first order creeks, are sentinel habitats for larger estuarine systems. They provide early warning of potential impacts, as they are the primary link between estuarine and upland habitats and serve as conduits for runoff from surrounding uplands (Lerberg et al. 2000; Holland et al. 2004; Washburn and Sanger 2011; Sanger et al. 2015). They are also recreationally, commercially, and aesthetically important which results in significant coastal population growth. Between 1970 and 2010, Southeast coastal counties had the greatest rate of population increase in the United States (The Nature Conservancy, 2016), with populations in North Carolina, South Carolina, and Georgia experiencing 59%, 83%, and 72% increases in population change, respectively, and projected population changes from 2010 to 2020 being 8% (NC), 17% (SC), and 9% (GA) (Crossett et al. 2013). Additionally, population models predict that by 2025 between 25 and 30 percent of coastal watersheds will be under development in the Southeast (Allen and Lu 2003). Due to this rapid development, the watersheds of these headwater creeks and their surrounding estuarine area is rapidly changing from coastal forested and agricultural land to suburban and urban developed areas at more than twice the rate of coastal population growth (Crossett et al. 2004).

Creeks chosen for this sampling effort represented a diverse set of watershed land-use classifications based on the classifications developed by Holland et al. (2004). Watershed land-use classifications were defined by percent

IC within the watershed; forested watersheds had <10% IC, suburban watersheds had >10% but <50% IC, and urban watersheds had >50% IC. In addition to the above classifications, a forested to suburban land-use class was defined for this study as watersheds with IC that increased over the 20-year period. Six creeks were chosen to represent this new class, which had watersheds that changed from forested watersheds (10% IC) in the early 1990s to suburban watersheds (10–50% IC) associated with the 2014/2015 sampling.

Much of the research involving headwater tidal creek health in the Southeast US was based on procedures developed for the TCP in 1994. The TCP initially targeted 28 tidal creeks in the summers of 1994 and 1995 under the Charleston Harbor Study (n = 28, Sanger et al. 1999a, b; Lerberg et al. 2000, Holland et al. 2004). Additionally, creeks were added to the TCP paradigm and others were resampled with the May River Study in the summer of 2002 (n = 6, Van Dolah et al. 2004), and with the Ecological Research Assessment and Prediction (ERAP) project during the summers of 2005 and 2006 (n = 19, Washburn and Sanger 2011; Sanger et al. 2015). In this study, 18 of the creeks identified in the previous studies were resampled in the summers of 2014 and 2015 (Fig. 1, Table 1). While tidal creek habitats experience environmental stressors year-round, sampling supporting the TCP has always occurred during July and August as the impact of stressors such as temperature-driven dissolved oxygen changes and changes in salinity is magnified during the hotter summer months (Hackney et al. 1976) which experience average highs between the high 80s and low 90s. These stressors are also often exacerbated by increased levels of impervious cover so differences between land use classes will be more apparent.

Impervious Cover Calculation

McHouell (2016) details the methodology related to the watershed delineations and land features. Briefly, an ~1000 m transect was drawn in the middle of the channel of each creek, using ArcGIS 10.1, to identify the headwater sampling area. The sampling area (transect) was divided into three reaches, with one primary sampling location randomly identified in the middle reach. Watersheds surrounding the creeks were delineated in ArcGIS 10.1 using United States Geological Survey (USGS) topographic maps and Light Detection and Ranging (LIDAR) imagery. The percent IC in each creek's watershed was obtained using the 2016 USGS National Land Cover Dataset (NLCD). Only the non-marine wetland area of each watershed was used to determine the IC which was adjusted by quadratic equation to ensure comparability with 1994/1995 data (McHouell 2016). The historical IC dataset used the closest time period with IC data available for 2001 and 2006 (NLCD), and a point sampling method in 1994/1995 (Lerberg 1997).

Sample Collection

Eighteen creeks from the TCP's historical sampling efforts were selected in a stratified random design based on land use. Each creek represents one of the four land use classes identified above. Sampling was divided over the course of two summers, with nine creeks selected for sampling during the summer of 2014 and the remaining nine sampled in the summer of 2015. Each creek was sampled around low tide with no water present during sediment collection. Sediment was collected at the identified primary site in the middle reach of each creek, at approximately one meter below mean high water. Multiple scrapes of the upper two

Fig. 1 The locations of the 18 creeks sampled in 2014 and 2015, identified by land use class

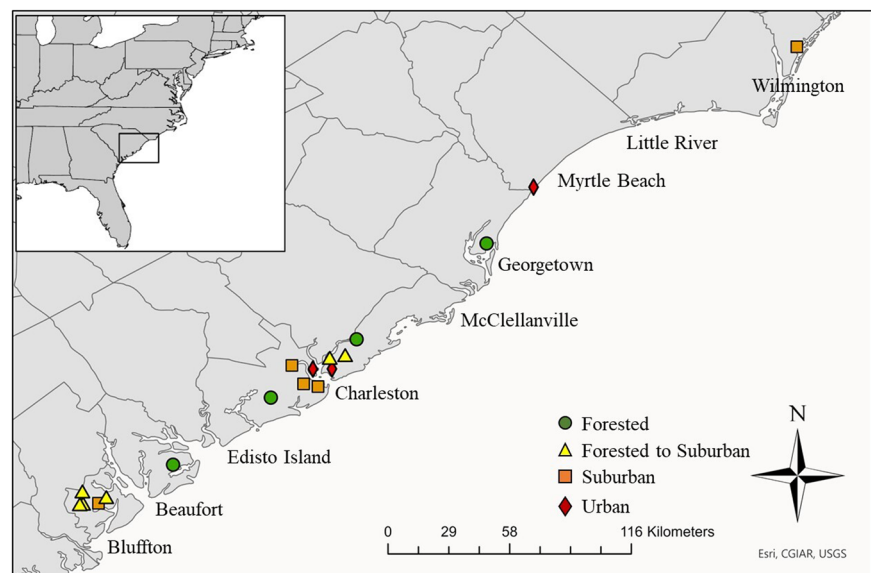


Table 1 Summary data for the 18 creeks sampled in the summers of 2014 and 2015

Creek name (Abbreviation)	Watershed land-use classification	County	State	GPS Coord.		WS (ha)	2016 IC (%)
				Lat.	Long.		
Deep (DP)	Forested	Charleston	SC	32.934	−79.750	64.9	2.93
Crabhaul (NI)	Forested	Georgetown	SC	33.346	−79.203	199.3	3.20
Long (LC)	Forested	Charleston	SC	32.686	−80.127	410.4	3.43
Village (VL)	Forested	Beaufort	SC	32.387	−80.552	629.7	3.71
Stoney (SC)	Forested to Suburban	Beaufort	SC	32.242	−80.954	2064.2	16.56
Sawmill (SW)	Forested to Suburban	Beaufort	SC	32.250	−80.833	600.2	18.94
Rose Dhu (RD)	Forested to Suburban	Beaufort	SC	32.245	−80.915	893.0	23.87
Horibeck (HB)	Forested to Suburban	Charleston	SC	32.868	−79.796	236.8	27.35
Okatee (OT)	Forested to Suburban	Beaufort	SC	32.278	−80.937	2410.6	32.09
Rathall (RT)	Forested to Suburban	Charleston	SC	32.856	−79.869	71.8	43.30
Parrot (PC)	Suburban	Charleston	SC	32.733	−79.921	133.5	27.38
Cross (CC)	Suburban	Charleston	SC	32.753	−79.981	310.5	39.09
Hewlitts (HL)	Suburban	New	NC	34.207	−77.863	458.8	44.30
Bulls (BL)	Suburban	Charleston	SC	32.819	−80.037	436.0	47.97
Heyward (HW)	Suburban	Beaufort	SC	32.246	−80.853	238.6	48.02
Murrells Inlet (MI)	Urban	Georgetown	SC	33.594	−78.996	456.3	56.97
Shem (SM)	Urban	Charleston	SC	32.814	−79.852	403.1	61.47
New Market (NM)	Urban	Charleston	SC	32.806	−79.945	196.9	72.59

Creek abbreviation, land-use class, general location, watershed size (WS), and 2016 watershed impervious cover (IC) level is identified for each creek

centimeters of exposed surface sediment were taken using a stainless-steel spoon and placed in a stainless-steel bowl. The sediment was homogenized in the bowl and portioned into a resealable plastic bag for sediment composition (i.e., sand, silt, clay, and total organic carbon contents), and clean contaminant sample jars (i.e., metals were collected in pre-cleaned plastic containers and organics in pre-cleaned glass jars). Samples were placed on ice until reaching the laboratory, where contaminant samples were frozen at -40°C and sediment composition samples were frozen at -12°C .

Sample Processing

A subsample of approximately ten grams from the sediment composition sample was removed for total organic carbon (TOC) analysis according to method 9060A (US EPA 2004). Total Organic Carbon for the nine creeks sampled in 2014 was analyzed at the South Carolina Department of Natural Resources (SC DNR) using a Perkin Elmer Model 2400 CHNS Analyzer. TOC for the nine creeks sampled in 2015, as well as duplicate samples from five creeks sampled in 2014, were analyzed by GEL Laboratories in Charleston, SC using a Dohrmann DC-190 Boat Sampler. The GEL duplicate 2014 TOC values were compared to the SC DNR values, with no significant differences between values, indicating the two laboratories produced comparable data.

Each sediment composition sample was also analyzed for percent sand, silt, and clay using a modified pipette method (Plumb 1981). Samples were thawed and homogenized then approximately 20 g of each sample was weighed and dispersed with 20 mL of sodium hexametaphosphate. The sample was wet-sieved with distilled water through a 63 μm sieve into a 1000 mL graduated cylinder. The material remaining in the sieve was considered sand and rinsed into a pre-weighed beaker. Once all silt and clay particles were sieved through, the cylinder was filled to 1000 mL with distilled water and homogenized by inverting the cylinder ten times. A 20 mL sample was then extracted at a depth of 20 cm and placed in a pre-weighed beaker for silt/clay determination. A second 20 mL sample was extracted at a specified amount of time later, depending on the temperature of the sample, at a depth of 5 cm to obtain a separate percent clay value and placed in its own beaker. All beakers were dried overnight and weighed the next day to determine the composition of the sediment sample; percent silt was determined by subtraction. Quality Assurance Quality Control (QAQC) was performed by a different processor who re-ran one random sample out of every ten samples. If the composition of sand and silt/clay differed by more than 10% between the original and the re-analyzed sample, the other nine samples in the batch were re-analyzed by the original processor.

Sediment contaminant samples were analyzed by the National Oceanic and Atmospheric Administration (NOAA)

Hollings Marine Laboratory in Charleston, SC. The contaminants targeted included 22 metals, 25 pesticides, 28 PAHs, 94 PCBs, and 14 PBDEs. Detailed methodology for both the historical and current datasets was similar to that provided by Kucklick et al. (1997), Long et al. (1998b), Balthis et al. (2012), and Chen et al. (2012). Briefly, samples were ground with anhydrous sodium sulfate (Na_2SO_4) to remove moisture, and then processed and extracted through an Accelerated Solvent Extractor 200 (ASE, Dionex Inc.). Interferences were removed by gel permeation chromatography (GPC) and solid phase extraction (SPE). Dry mass organic contaminant values were quantified with gas chromatography–mass spectrometry (GCMS, Agilent 6890/5972) using selected ion monitoring. For metals, sediment was dried and ground, followed by microwave nitric acid digestion. Digested samples were diluted, and metals analysis was completed by inductively coupled plasma mass spectrometry (ICPMS). Mercury concentrations were quantified by direct combustion (DMA80; Milestone, Inc.). Various QAQC samples were included with each batch of samples including: (1) duplicate samples, (2) blank samples, (3) reagent-spiked samples, or (4) certified National Institute of Science and Technology standard reference material (SRM) samples.

Data Summary and Analysis

Data management

All data collected for the TCP was archived in a relational Microsoft Access™ database managed by the SC DNR. All historical sediment contaminant and composition data from the projects that occurred between 1994 and 2006 were compiled into one dataset (referred to as the historical dataset). The historical dataset used for this analysis contains data for 43 unique tidal creeks. Six of the 43 unique creek systems were sampled in 2005/2006 as well as 1994/1995, resulting in a total historical sample size of 49 creeks.

Contaminant concentrations below the method detection limit (MDL) were converted to zero for statistical analysis. If the frequency of detection for a metal or compound was below 50%, no statistical analyses were conducted. Two metals, several PAH compounds, and numerous pesticide, PCB, and PBDE compounds experienced low frequencies of detection. Thus, analyses were conducted for the individual metals and PAHs with 50% or greater detection, total PAH, total low molecular weight (LMW) PAH, total high molecular weight (HMW) PAH, and total values for DDT, PCB, and PBDE, although data for the individual compounds can be found in the supplemental information (Online Resource 1). Total DDT was calculated by summing the concentrations of six DDT metabolites, total PCB

summed the concentrations of 94 congeners and total PBDE summed 14 congeners.

In the regression analysis, total $\text{PBDE}_{(-209)}$ indicates a total PBDE metric calculated without PBDE 209 as this analyte was not measured for until the 2014/2015 sampling. For analysis of covariance (ANCOVA) and analysis of variance (ANOVA) on current data, total values include PBDE 209. Chemical contamination was assessed using the sediment quality guidelines Effects Range Median (ERM) and Effects Range Low (ERL) (Long et al. 1995). A mean Effects Range Median Quotient (ERM-Q) was calculated as an integrative assessment of potential contaminant effects that included metals ($n = 8$), PAHs ($n = 13$), Total DDT, and Total PCBs. There are currently no identified ERLs or ERM-Qs for PBDEs to include the class in overall mean ERM-Q. Mean ERM-Q was calculated by dividing each concentration by its ERM, summing the results, then dividing by the total number of compounds included in the calculation (Long et al. 1998a). Separate mean ERM-Qs for metals and PAHs were also calculated. DDT only has an ERM for one compound and Total DDT, while PCBs only have an ERM for Total PCB, so mean ERM-Q was not calculated for these classes. ERM-Qs were calculated in an effort to normalize contaminant results to the levels at which biological effects are probable.

Data analysis

Univariate data analysis was conducted using the R statistical software version 3.4.2 in R Studio version 1.1.383 (R Core Team 2013). Normality of residuals was examined using the Shapiro-Wilks test. In cases where the data failed to meet the assumption of normality, the data were transformed using $\log_{10}(x)$ or $\log_{10}(x + 1)$. Analyses were considered significant where p -values ≤ 0.05 and marginally significant where p -values were > 0.05 and < 0.10 . Linear regression was used to evaluate sediment composition against percent watershed IC and contaminant class concentrations against percent clay or percent TOC and percent watershed IC. Both historical and current contaminant data were regressed against their respective watershed IC levels and compared. The effect of clay or TOC content among the land-use categories was assessed using one-way ANCOVA. Clay content was identified as a covariate for metal concentrations while TOC content was identified as a covariate for PAHs, total DDT, total PCB, and total PBDE. Contaminants tend to bind to clay or organic carbon, and it is expected that in creeks with high clay or high TOC levels there will be higher contaminant levels (Sanger et al. 2008). If a covariate was found to be significant at $\alpha = 0.05$, then a one-way ANCOVA was performed to control the effect of the covariate. If a covariate was not found to be significant, a one-way ANOVA was performed to evaluate the potential

differences in sediment contaminants and metrics among land-use categories. In all cases, Least Squares Means were used for pairwise comparisons to identify which classifications differed significantly from each other, if any.

The number of individual tests conducted in this study has the potential of increasing the likelihood of obtaining false positives, Type I error. To account for this, Rice (1989) suggested using the Bonferroni adjustment (1) when two or more tests are conducted, and the p-values determine where statistically significant differences occur, or (2) when two or more tests are conducted to reject a common null hypothesis and the null hypothesis would be rejected if only some but not all tests were significant. The Bonferroni adjustment deflates the α by dividing α by the number of tests performed within a group, thus reducing the chance for Type I error by making p-values more conservative (Rice 1989; Benjamini and Hochberg 1995; Perneger 1998). A primary argument in the literature against using this adjustment is that by reducing the chance for Type I error one will also increase the chance for Type II error (false negatives) (Perneger 1998; Cabin and Mitchell 2000; Moran 2003). The Bonferroni adjustment was not utilized in this study for the following two reasons. An α of 0.05 in ecological studies, such as this one, is already conservative considering the variability that occurs within these systems. Also, the goal of the TCP and this study was to assess the effects of coastal development on sediment quality using an ecosystem approach with a large number of metrics. Therefore, the focus was on patterns of significance among groups of tests as a weight of evidence approach instead of relying on one significant test to reject the null hypothesis.

Multivariate analyses were conducted using PRIMER (version 7; Clarke and Gorley 2015). Data were individually $\log_{10}(x)$ transformed then normalized in PRIMER before analysis. Total DDT, total PCB, total PBDE, and the individual metals and PAHs with ERM values were combined into one dataset for this analysis. Hierarchical cluster modeling (CLUSTER), similarity profile (SIMPROF) analysis using Euclidian distance, and non-metric multidimensional scaling (nMDS) analyses were performed to assess the similarity among creeks for contaminant concentrations. The sediment composition parameters clay and TOC were overlaid as vectors to determine if they had an effect on the distribution of sediment contaminants across creeks. Vector direction indicates the direction in which values increase and vector length reflects the strength of the factor in that direction; the circle indicates a multiple correlation of one. An unordered Analysis of Similarity (ANOSIM) was also performed to test significant difference between land-use classifications. Due to highly variable environmental conditions, a p-value of 0.1 was used for these analyses.

Metal enrichment was determined in each creek following methods described in Schropp et al. (1990) and Hanson

et al. (1993) by first regressing forested creek metal concentrations against respective aluminum values and calculating upper and lower 95% prediction intervals. Using a linear regression approach, only metals which were statistically significantly related to aluminum were used in this analysis. To provide a more robust sample of reference creeks, all historical and current reference/forested creek data were used to create the prediction intervals ($n = 24$). Metal concentrations for the current developed creeks were overlaid on the reference regression, and any concentrations exceeding the reference upper 95% prediction interval were considered enriched beyond natural levels.

The number of times a concentration exceeded the sediment quality guideline of ERL and ERM in each creek were quantified. Concentrations above the ERL and below the ERM fall in a “possible-effects” range in which toxic contaminant effects could be occasional, and concentrations above the ERM fall in a “probable-effects” range in which effects could be frequent but exceeding an ERL or ERM value does not instantly qualify sediment as being toxic (Long et al. 1995). The United States Environmental Protection Agency (US EPA) in their National Coastal Condition Report (2012) considers sediment quality with respect to chemical contamination as “poor” if there was at least one ERM exceedance, “fair” if no ERMs were exceeded but five or more ERLs were exceeded, and “good” if < five ERLs were exceeded.

Finally, a paired analysis for clay and TOC content, total DDT, total PCB, total PAH, total LMW PAH, total HMW PAH, ERM-Q, mean metal ERM-Q, and mean PAH ERM-Q was conducted on data from the 11 headwater tidal creeks in the Southeast which had data from two time periods over a 20-year span (1994/1995 and 2014/2015). Land use of these creeks was classified as forested ($n = 3$), forested to suburban ($n = 3$), suburban ($n = 3$), and urban ($n = 2$). Total PBDE was not included in the paired analysis as PBDEs were not analyzed for during the 1994/1995 sampling. All parameters for the 11 creeks were first individually graphed to qualitatively determine changes over time. Paired t-tests were then conducted to identify changes over time across all land-use classes.

Results

Trace Metal Enrichment

Trace metal enrichment based on aluminum content was evaluated, and eight trace metals (cadmium [Cd], copper [Cu], lead [Pb], lithium [Li], manganese [Mn], mercury [Hg], selenium [Se], and thallium [Tl]) were found to be statistically significantly related to aluminum levels in forested creeks (Table 2, Fig. 2). Concentrations of

Table 2 List of trace metals which exhibit statistically significant relationships with aluminum in forested creeks

Creek	Cd	Cu	Hg	Li	Mn	Pb	Se	Tl	Total
Deep				■					1
Crabhaul			■						1
Long									0
Village									0
Stoney								■	1
Sawmill	■								1
Rose Dhu									0
Horlbeck									0
Okatee									0
Rathall								■	1
Parrot	■			■				■	4
Cross									0
Hewlitts	■		■				■		3
Bulls	■		■	■				■	5
Heyward									0
Murrells Inlet									0
Shem	■	■	■						3
New Market	■	■	■	■		■	■		6

The shaded squares represent the instances in which a metal was deemed enriched by exceeding the upper 95% prediction interval for forested creek concentrations

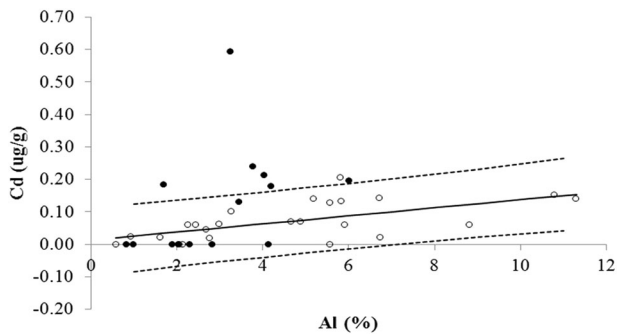


Fig. 2 A metal enrichment analysis example. Reference forested creeks are identified by open circles and developed creeks sampled in 2014/2015 are by solid circles. A solid regression line and dashed 95% upper and lower prediction intervals are overlain. Developed creeks above the upper 95% prediction interval have sediments enriched with cadmium relative to reference concentrations

cadmium, copper, lead, manganese, selenium, and thallium were found to be enriched in developed creeks. Results of this data indicated that creeks categorized as forested and forested to suburban only had up to one of these metals considered to be enriched, based on concentrations exceeding the upper 95% prediction interval for forested creek concentrations. The suburban creeks had up to five elements that were identified as enriched, and the urban creeks had up to six enriched metals. One creek system, Murrells Inlet Creek (urban), did not exhibit any enrichment based on this data.

ERL and ERM Exceedances

The number of ERL and ERM exceedances for metals, PAHs, DDT, and total PCB were also calculated (Table 3). Concentrations for the metals cadmium, chromium, and nickel did not exceed either the ERL or ERM values in any of the 18 creeks. Arsenic levels exceeded the ERL value in five creeks: Crabhaul, Parrot, Hewlitts, Shem, and New Market. Zinc levels exceeded the ERM value of 410 ug/g in Crabhaul (539 ug/g), Long (470 ug/g), Stoney (545 ug/g), Rose Dhu (419 ug/g), Heyward (416 ug/g), and Shem (505 ug/g) creeks and the ERL value of 150 ug/g in Cross, Hewlitts, and New Market creeks. The ERL value for mercury of 0.15 ug/g was exceeded in Shem (0.16 ug/g) and New Market (0.32 ug/g) creeks, while New Market also exhibited ERL (34 ug/g and 47 ug/g, respectively) exceedances for copper (62 ug/g) and lead (217 ug/g) (lead nearly exceeded its ERM value of 218 ug/g).

Exceedances for PAH concentrations were primarily observed above ERLs in Shem and New Market creeks (nine and twelve ERL exceedances, respectively), which includes total PAH. Total HMW PAH exceeded its ERM (9,600 ng/g) in New Market Creek (15,018 ng/g). The creeks Bulls and Heyward Cove also exceeded total HMW PAH ERL (1,700 ng/g) concentrations at 3,214 ng/g and 1,812 ng/g, respectively. Exceedances for individual compounds only occurred in the urban creeks New Market and Shem. The only individual DDT metabolite to have an ERL

Table 3 The number of Effects Range Median (ERM) and Effects Range Low (ERL) exceedances for each creek sampled in 2014 and 2015 and where the exceedances occurred

Creek	Mean ERM-Q	Total ERM exceedances	Total ERL exceedances	Metals	PAHs	DDT	Total PCB
Deep	0.0156	0	0	0	0	0	0
Crabhaul	0.0761	1	1	2	0	0	0
Long	0.0690	1	2	1	0	2	0
Village	0.0039	0	0	0	0	0	0
Stoney	0.0682	1	0	1	0	0	0
Sawmill	0.0111	0	0	0	0	0	0
Rose Dhu	0.0480	1	0	1	0	0	0
Horlbeck	0.0247	0	0	0	0	0	0
Okatee	0.0115	0	0	0	0	0	0
Rathall	0.0307	0	0	0	0	0	0
Parrot	0.0385	0	2	1	0	1	0
Cross	0.0315	0	2	1	0	1	0
Hewlitts	0.0685	0	4	2	0	2	0
Bulls	0.0763	0	2	0	1	1	0
Heyward	0.0787	1	2	1	1	1	0
Murrells Inlet	0.0122	0	0	0	0	0	0
Shem	0.1911	1	12	3	9	1	0
New Market	0.3935	2	18	5	13	1	1

Bold values indicate where ERM exceedances occurred

and ERM value is 4,4'-DDE; its ERL value (2.2 ng/g) was exceeded in Long (4.66 ng/g), Hewlitts (3.86 ng/g), and New Market (11.64 ng/g) creeks. Total DDT, however, exceeded the ERL value (1.58 ng/g) in eight creeks: Long (7.09 ng/g), Parrot (1.75 ng/g), Cross (1.91 ng/g), Hewlitts (5.84 ng/g), Bulls (1.68 ng/g), Heyward Cove (1.69 ng/g), Shem (2.80 ng/g), and New Market (21.30 ng/g). As for total PCB, only the ERM (180 ng/g) was exceeded in New Market Creek (186 ng/g).

Regression Analysis

Currently, neither clay nor TOC content exhibited statistically significant relationships with watershed IC. Historically, clay content did not exhibit a relationship although TOC content did (Table 4). Regressing sediment clay content against individual metal concentrations in the creeks resulted in significant relationships for aluminum, arsenic, beryllium, cobalt, chromium, copper, iron, lithium, mercury, manganese, nickel, lead, selenium, thallium, uranium, and vanadium. Sediment TOC content was regressed against individual PAH analytes (Table 4). Relationships were statistically significant or marginally significant in all cases but two, benzo(b)fluoranthene and pyrene; these compounds exhibited variable concentrations across creeks, with many less developed creeks having concentrations similar to more developed creeks. Total PAH and total HMW PAH were significantly related to TOC while total LMW PAH was not.

Total PAH was driven by HMW PAH concentrations, thus similar relationships for total PAH and total HMW PAH were expected. Sediment TOC content was also significantly related to total DDT, total PCB, and total PBDE.

Concentrations of the metals cadmium, copper, lead, and uranium were significantly related to watershed IC both currently and historically (Table 4). Chromium and tin were related currently but not historically, whereas mercury, selenium, and zinc were not related currently but were historically. Finally, concentrations of aluminum, arsenic, barium, beryllium, cobalt, iron, lithium, manganese, nickel, thallium, and vanadium were not related to IC currently or historically. All individual PAHs (except benzo(a)fluoranthene and retene), total PAH, total LMW PAH, total HMW PAH, total DDT, and total PCB were significantly related to watershed IC in the regression analysis, with increasing concentrations observed in conjunction with increasing IC levels. These relationships were similar to those observed historically (Table 4, Fig. 3). Total PBDE was also related to IC currently and total PBDE₍₋₂₀₉₎ was related to watershed IC both currently and historically with the current regression for total PBDE₍₋₂₀₉₎ nearly identical to that for the historical data. As for ERM-Q quotients, only mean ERM-Q and mean PAH ERM-Q were significantly related to watershed IC currently; in the historical analysis, all three quotients were related (Table 4, Fig. 4). The urban creeks New Market and Shem had higher contamination quotients than all other creeks. The forested creek Crabhaul

Table 4 Linear regression analysis results for current (2014/2015) and historical datasets (1994–2006) for sediment composition, individual contaminants, and Effects Range Median Quotients versus impervious cover (IC), clay, and TOC

Parameter	IC (current)			IC (historical)			Clay (current)		
	R ²	p-value	slope	R ²	p-value	slope	R ²	p-value	slope
Clay (%)	0.03	0.524	0.116	0.03	0.242	−0.172			
TOC (%)	0.10	0.207	0.059	0.09	0.040	0.025			
ERM-Qs									
Mean ERM-Q*	0.24	0.037	0.011	0.33	<0.001	0.010			
Mean Metal ERM-Q	0.10	0.197	0.001	0.25	<0.001	0.003			
Mean PAH ERM-Q**	0.45	0.002	0.001	0.34	<0.001	0.001			
Metals (% or ug/g dry)									
Aluminum** (%)	0.06	0.346	0.002	0.04	0.145	−0.002	0.57	<0.001	0.009
Arsenic**	0.18	0.083	0.006	0.01	0.517	0.001	0.51	<0.001	0.015
Barium*	0.03	0.467	0.003	0.13	0.126	0.003	0.05	0.354	0.005
Beryllium	0.09	0.217	0.002	0.02	0.523	0.003	0.62	<0.001	0.009
Cadmium**	0.38	0.007	0.004	0.41	<0.001	0.002	0.14	0.126	0.004
Chromium**	0.32	0.015	0.005	0.03	0.228	0.004	0.58	<0.001	0.009
Cobalt*	0.16	0.099	0.003	0.06	0.294	0.003	0.62	<0.001	0.009
Copper**	0.43	0.003	0.011	0.10	0.031	0.008	0.38	0.006	0.014
Iron (%)	0.04	0.418	0.010	0.00	0.819	−0.001	0.40	0.005	0.043
Lead*	0.43	0.003	0.012	0.25	<0.001	0.008	0.33	0.012	0.014
Lithium*	0.17	0.090	0.011	0.10	0.181	0.004	0.55	<0.001	0.027
Manganese*	0.09	0.223	0.004	0.00	0.730	−0.001	0.55	<0.001	0.012
Mercury*	0.19	0.074	0.009	0.14	0.008	0.007	0.45	0.002	0.019
Nickel*	0.13	0.138	0.005	0.00	0.850	0.000	0.61	<0.001	0.016
Selenium**	0.14	0.128	0.002	0.08	0.048	0.001	0.56	<0.001	0.004
Thallium	0.18	0.078	0.003	0.03	0.441	0.001	0.65	<0.001	0.007
Tin*	0.33	0.013	0.011	0.00	0.917	−0.001	0.01	0.676	0.003
Uranium*	0.28	0.025	0.006	0.37	0.006	0.005	0.51	0.001	0.011
Vanadium*	0.18	0.079	0.003	0.03	0.508	0.001	0.61	<0.001	0.008
Zinc**	0.01	0.659	0.003	0.37	<0.001	0.010	<0.001	0.941	0.001
PAHs (ng/g dry)							TOC (%) (current)		
1-Methylphenanthrene**	0.58	<0.001	0.020	0.53	<0.001	0.022	0.27	0.028	0.072
Acenaphthylene**	0.33	0.013	0.014	0.52	<0.001	0.022	0.17	0.089	0.052
Anthracene**	0.66	<0.001	0.023	0.55	<0.001	0.028	0.22	0.050	0.070
Benzo(a)anthracene**	0.66	<0.001	0.031	0.50	<0.001	0.029	0.28	0.025	0.106
Benzo(a)pyrene**	0.69	<0.001	0.032	0.53	<0.001	0.031	0.30	0.018	0.111
Benzo(b)fluoranthene**	0.70	<0.001	0.037	0.50	<0.001	0.030	0.16	0.105	0.091
Benzo(e)pyrene**	0.70	<0.001	0.032	0.51	<0.001	0.028	0.33	0.013	0.116
Benzo(g,h,i)perylene**	0.73	<0.001	0.036	0.49	<0.001	0.028	0.19	0.067	0.098
Benzo(k+j)fluoranthene**	0.72	<0.001	0.034	0.45	<0.001	0.026	0.27	0.026	0.109
Benzo(a)fluoranthene**	0.69	<0.001	0.027	x	x	x	0.21	0.057	0.079
Chrysene+Triphenylene**	0.71	<0.001	0.034	0.52	<0.001	0.029	0.30	0.019	0.115
Dibenz(a,h)anthracene**	0.72	<0.001	0.029	0.57	<0.001	0.026	0.23	0.042	0.088
Dibenzothiophene**	0.50	0.001	0.015	0.24	0.013	0.010	0.43	0.003	0.074
Fluoranthene**	0.74	<0.001	0.034	0.40	<0.001	0.027	0.27	0.027	0.108
Fluorene**	0.60	<0.001	0.018	0.41	<0.001	0.019	0.35	0.010	0.074
Indeno(1,2,3-cd)pyrene**	0.61	<0.001	0.030	0.62	<0.001	0.035	0.36	0.008	0.123
Perylene**	0.57	<0.001	0.019	0.51	<0.001	0.024	0.57	<0.001	0.103

Table 4 (continued)

Parameter	IC (current)			IC (historical)			Clay (current)		
	R^2	p -value	slope	R^2	p -value	slope	R^2	p -value	slope
Phenanthrene**	0.70	<0.001	0.033	0.50	<0.001	0.025	0.20	<i>0.060</i>	0.094
Pyrene**	0.79	<0.001	0.040	0.51	<0.001	0.028	0.15	0.110	0.093
Retene*	0.14	0.132	0.009	x	x	x	0.53	0.001	0.091
Total (ng/g dry)									
Total PAH**	0.64	<0.001	0.028	0.42	<0.001	0.027	0.32	0.013	0.115
Total LMW PAH**	0.65	<0.001	0.035	0.38	<0.001	0.022	0.14	0.130	0.070
Total HMW PAH**	0.64	<0.001	0.032	0.45	<0.001	0.028	0.33	0.013	0.118
Total DDT**	0.27	0.027	0.009	0.36	<0.001	0.010	0.28	0.025	0.046
Total PCB**	0.34	0.011	0.013	0.25	<0.001	0.012	0.37	0.007	0.075
Total PBDE**	0.42	0.004	0.012	x	x	x	0.61	<0.001	0.079
Total PBDE ₍₋₂₀₉₎ **	0.27	0.029	0.005	0.46	0.001	0.005	0.62	<0.001	0.077

Significant ($p < 0.05$) relationships are identified in bold, marginally significant ($p > 0.05$ and < 0.1) are identified in italics. Analytes marked with * have been $\log_{10}(x)$ transformed and analytes marked with ** have been $\log_{10}(x + 1)$ transformed. An “x” indicates less than half of the creeks had detected concentrations, so it was not included in analysis

had mean ERM-Q and mean metal ERM-Q values similar or greater than those found in suburban creeks, in part due to its high levels of zinc and total DDT.

ANCOVA and ANOVA Analyses

Average percent clay in the eighteen creeks ranged from 22–30%, with no significant differences between watershed land-use classes ($p = 0.918$). Average percent TOC ranged from 2–7%, with no statistically significant differences between watershed land-use classes ($p = 0.436$). Despite no significant difference, creeks in urban watersheds had the highest average TOC content followed by suburban, forested, then forested to suburban (Table 5).

Clay content was a significant covariate for 16 of the metals analyzed. After the variability associated with the clay content was removed, there were no significant differences among land-use classes for metals except for copper and uranium (Table 5). Concentrations of copper and uranium in urban creeks were significantly greater than those in forested and forested to suburban creeks. One-way ANOVA models were conducted for barium, tin, and zinc, and no significant differences were observed among the land-use classes. Despite the lack of significant differences among land-use classes for most metals, there was an overall trend where urban and suburban creeks had higher metal concentrations than forested and forested to suburban classes within clay samples.

Total organic carbon content was a significant covariate for 19 of the 20 PAHs analyzed, as well as for total PAH, total LMW PAH, and total HMW PAH (Table 5). After the variability associated with the TOC content was removed, significant differences among watershed classifications were

observed for all analytes except total LMW PAH. In nearly every case, concentrations in forested creeks were significantly lower than those in both suburban and urban creeks. In 13 of these cases, concentrations in forested to suburban creeks were also significantly different than those in urban creeks, but not different than those in suburban creeks. Overall, the urban and suburban creeks had higher PAH concentrations than those with forested and forested to suburban classes. Total DDT and total PCB did not exhibit any significant differences among land-use classes once the variability associated with TOC was removed. The overall trend showed forested and forested to suburban creeks with lower concentrations than suburban and urban creeks (Table 5). Differences among land-use classes did occur for total PBDE, with forested creek concentrations found to be significantly lower than all other land uses.

One-way ANOVA models were conducted for mean ERM-Q, mean metal ERM-Q, and mean PAH ERM-Q and no significant differences among land-use classes were observed for mean ERM-Q or mean metal ERM-Q (Table 5). However, mean PAH ERM-Q values in urban creeks were significantly higher than those in forested, forested to suburban, and suburban creeks, which were all similar to each other. Overall, the trend observed for all three quotients was that forested creeks had the lowest values, with forested to suburban having the next lowest values, then suburban, and finally urban creeks with the highest values (Fig. 4).

nMDS and ANOSIM

The nMDS plot showed a clear pattern of contaminant distribution, with the forested and forested to suburban

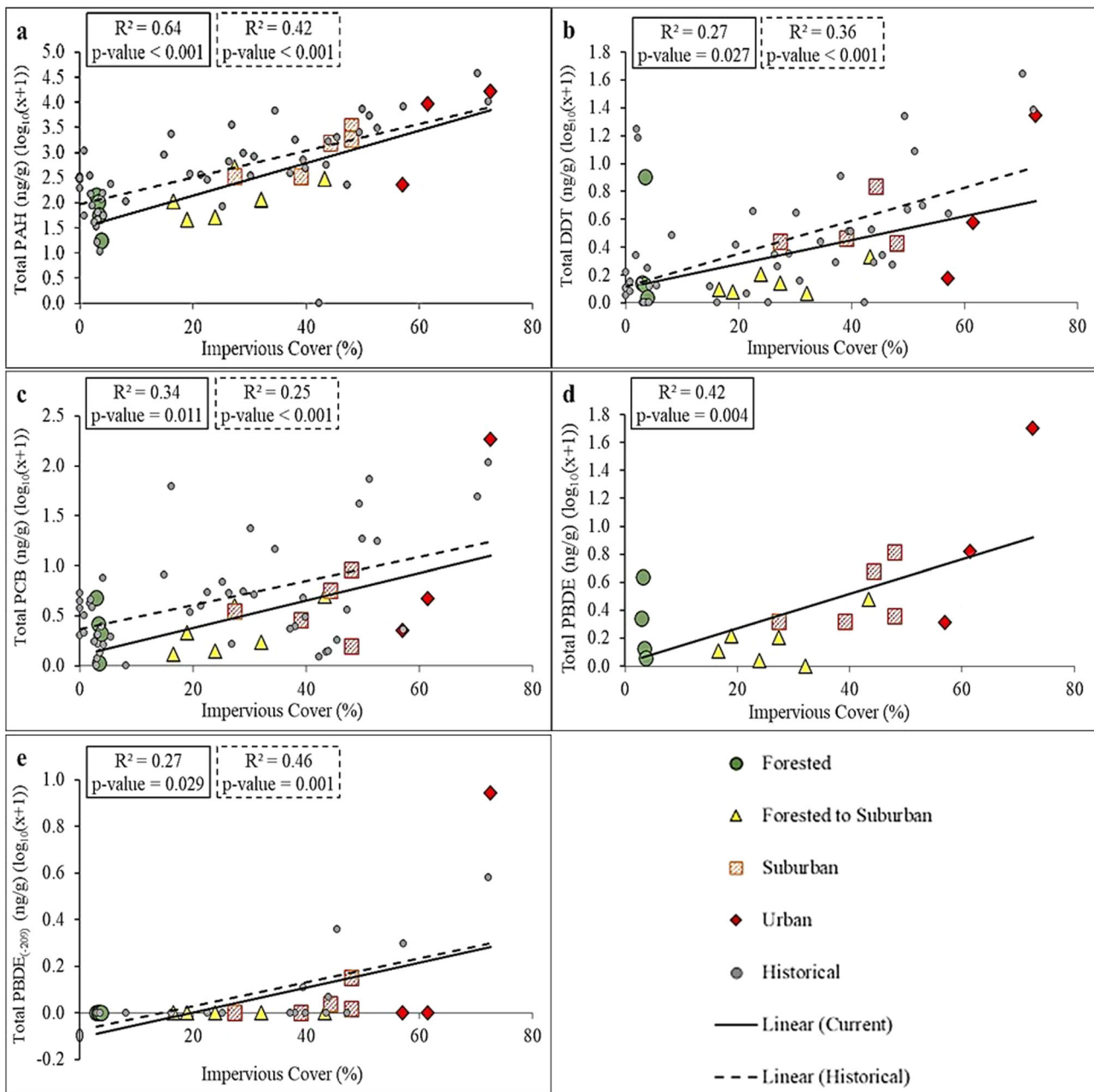


Fig. 3 Regression models for (a) total PAH, (b) total DDT, (c) total PCB, (d) total PBDE, and (e) total PBDE₍₋₂₀₉₎ vs. watershed impervious cover for current (2014/2015) and historical (1994–2006) datasets

creeks on the left side of the plot and the suburban and urban creeks on the right side (Fig. 5). Both clay and TOC content seemed to be driving some of the horizontal distribution. With the exception of Murrells Inlet Creek (MI), SIMPROF analysis identified significant groupings among creeks with similar impervious cover and contaminant levels, with New Market Creek isolated. At a significance level of 0.1, results from the ANOSIM showed significant differences between the forested class and suburban class (p-value = 0.0159), as well as the forested class and urban

class (p-value = 0.0571), with R values of 0.53 and 0.41, respectively (Table 6). The forested to suburban class was also significantly different from the suburban class (R = 0.30, p-value = 0.0498) and the urban class (R = 0.43, p-value = 0.0714).

Paired Analysis

Impervious cover, clay content, and total LMW PAH concentrations were the only parameters which exhibited

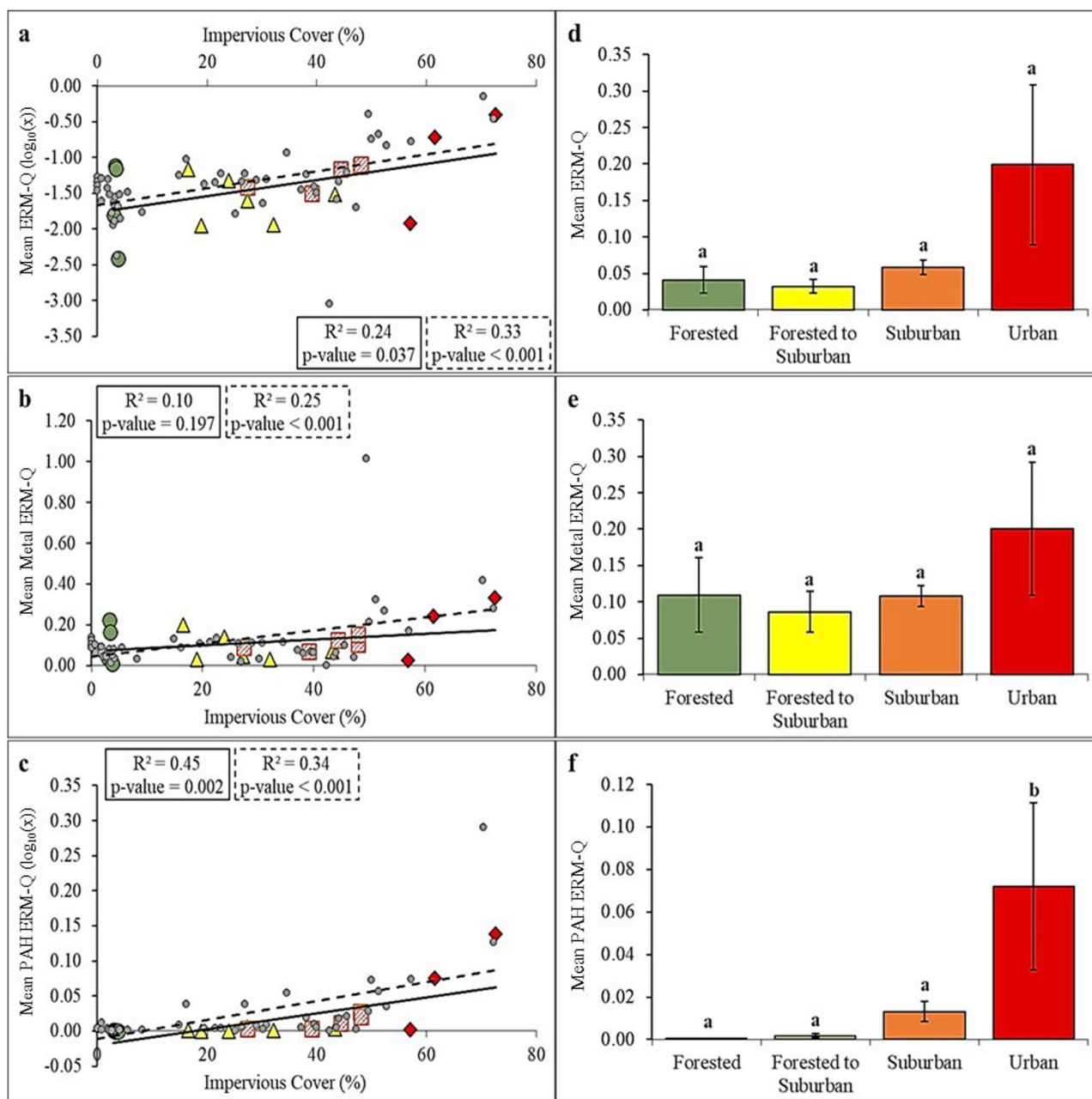


Fig. 4 Regression models (a–c) and mean values by land-use class (\pm SE) (d–f) for mean ERM-Q, mean metal ERM-Q, and mean PAH ERM-Q. Regression models for the current (2014/2015) and historical

(1994–2006) datasets. Lowercase letters indicate similarities and differences in the ANOVA/ANCOVA analyses

significant changes in their values over time, therefore a table displaying the temporal trend per creek over the 20-year period was chosen to describe changes instead (Table 7). Impervious cover increased significantly in all developed creeks and stayed consistent in forested creeks ($p = 0.004$). Clay content decreased significantly in all 11 creeks ($p = 0.001$), whereas TOC content exhibited variable changes. Increases in mean ERM-Q and mean metal ERM-Q in the forested creeks were primarily driven by high zinc

levels, while mean PAH ERM-Q remained similar to historical values in forested creeks. Increases in total PAH were linked to increases in total HMW PAHs, as overall, total LMW PAHs decreased significantly ($p = 0.048$). However, when forested creeks were removed from analysis, total LMW PAH did not exhibit a significant change over time ($p = 0.231$). Interestingly, total DDT increased in most of the developed creeks and one forested creek. Total PCB did not exhibit any consistent pattern of change across creeks.

Table 5 Analysis of Covariance (ANCOVA) and Analysis of Variance (ANOVA) results identifying differences among land-use classes

Parameter	Model R^2	Model p -value	Land-use class p -value	Covariate p -value	Land-use class LS means
Clay (%)	0.03	0.918	na	na	F ^a U ^a F ^a S ^a S ^a
TOC (%)	0.17	0.436	na	na	F ^a S ^a F ^a S ^a U ^a
ERM-Qs					
Mean ERM-Q*	0.24	0.270	na	na	F ^a F ^a S ^a S ^a U ^a
Mean Metal ERM-Q	0.19	0.390	na	na	F ^a S ^a F ^a S ^a U ^a
Mean PAH ERM-Q**	0.53	0.012	na	na	F^aF^aS^aS^aU^a
Metals (% or ug/g dry)				Clay (%)	
Aluminum** (%)	0.61	0.011	0.718	0.001	F ^a S ^a F ^a U ^a S ^a
Arsenic**	0.66	0.004	0.169	0.001	F ^a S ^a F ^a S ^a U ^a
Barium*	0.12	0.615	na	na	F ^a U ^a F ^a S ^a S ^a
Beryllium	0.66	0.004	0.679	<0.001	F ^a U ^a F ^a S ^a S ^a
Chromium**	0.77	<0.001	0.039	<0.001	F ^a F ^a S ^a S ^a U ^a
Cobalt*	0.67	0.004	0.593	<0.001	F ^a F ^a S ^a S ^a U ^a
Copper**	0.69	0.003	0.024	0.001	F^aF^aS^aS^a.b.U^a
Iron (%)	0.48	0.056	0.589	0.007	F ^a S ^a F ^a S ^a U ^a
Lead*	0.63	0.008	0.051	0.005	F ^a F ^a S ^a S ^a U ^a
Lithium*	0.71	0.002	0.120	<0.001	F ^a S ^a F ^a S ^a U ^a
Manganese*	0.62	0.009	0.493	0.001	F ^a F ^a S ^a U ^a S ^a
Mercury*	0.56	0.022	0.408	0.003	F ^a S ^a F ^a S ^a U ^a
Nickel*	0.69	0.003	0.363	<0.001	F ^a S ^a F ^a S ^a U ^a
Selenium**	0.72	0.002	0.118	<0.001	F ^a S ^a F ^a U ^a S ^a
Thallium	0.76	0.001	0.140	<0.001	F ^a F ^a S ^a U ^a S ^a
Tin*	0.28	0.120	na	na	F ^a F ^a S ^a S ^a U ^a
Uranium*	0.77	<0.001	0.020	<0.001	F^aF^aS^aS^a.b.U^a
Vanadium*	0.72	0.002	0.216	<0.001	F ^a F ^a S ^a U ^a S ^a
Zinc**	0.03	0.927	na	na	F ^a S ^a F ^a U ^a S ^a
PAHs (ng/g dry)				TOC (%)	
Acenaphthylene**	0.26	0.217	na	na	F ^a F ^a S ^a S ^a U ^a
Anthracene**	0.62	0.010	0.022	0.017	F^aF^aS^a.b.S^a.b.U^a
Dibenzothiophene**	0.71	0.002	0.030	0.001	F^aF^aS^a.b.S^a.b.U^a
Fluorene**	0.64	0.007	0.047	0.004	F^aF^aS^a.b.S^a.b.U^a
1-Methylphenanthrene**	0.58	0.016	0.055	0.013	F^aF^aS^a.b.S^a.b.U^a
Benzo(a)anthracene**	0.69	0.003	0.010	0.005	F^aF^aS^a.b.S^a.b.U^a
Benzo(a)fluoranthene**	0.65	0.006	0.012	0.016	F^aF^aS^a.b.S^a.b.U^a
Benzo(a)pyrene**	0.74	0.001	0.004	0.002	F^aF^aS^a.b.S^a.c.U^a
Benzo(b)fluoranthene**	0.69	0.003	0.004	0.024	F^aF^aS^a.b.S^a.c.U^a
Benzo(e)pyrene**	0.78	<0.001	0.002	0.001	F^aF^aS^a.b.S^a.c.U^a
Benzo(g,h,i)perylene**	0.72	0.001	0.003	0.010	F^aF^aS^a.b.S^a.c.U^a
Benzo(k+j)fluoranthene**	0.75	0.001	0.002	0.002	F^aF^aS^a.b.S^a.c.U^a
Chrysene+Triphenylene**	0.75	0.001	0.003	0.002	F^aF^aS^a.b.S^a.c.U^a
Dibenz(a,h)anthracene**	0.72	0.001	0.004	0.006	F^aF^aS^a.b.S^a.c.U^a
Fluoranthene**	0.74	0.001	0.003	0.003	F^aF^aS^a.b.S^a.c.U^a
Indeno(1,2,3-cd)pyrene**	0.74	0.001	0.007	0.001	F^aF^aS^a.b.S^a.c.U^a
Perylene**	0.83	<0.001	0.006	<0.001	F^aF^aS^a.b.S^a.b.U^a
Phenanthrene**	0.63	0.008	0.015	0.019	F^aF^aS^a.b.S^a.b.U^a
Pyrene**	0.73	0.001	0.001	0.018	F^aF^aS^a.b.S^a.c.U^a
Retene*	0.60	0.012	0.535	0.001	F ^a U ^a S ^a F ^a S ^a

Table 5 (continued)

Parameter	Model R^2	Model p -value	Land-use class p -value	Covariate p -value	Land-use class LS means
Total (ng/g dry)					
Total PAH*	0.76	0.001	0.003	0.001	F^aFS^{a,b}S^{b,c}U^c
Total LMW PAH**	0.60	0.017	0.021	<i>0.060</i>	F^aFS^{a,b}S^bU^b
Total HMW PAH*	0.77	<0.001	0.003	<0.001	F^aFS^{a,b}S^{b,c}U^c
Total DDT**	0.45	<i>0.081</i>	0.301	0.024	FS ^a F ^a S ^a U ^a
Total PCB**	0.52	0.039	0.324	0.007	F ^a FS ^a S ^a U ^a
Total PBDE**	0.78	<0.001	0.048	<0.001	F^aFS^{a,b}S^{a,b}U^b

Abbreviations are forested (F), forested to suburban (FS), suburban (S), and urban (U). Abbreviations are in order of increasing concentrations. Superscripts denote similarities or statistically significant differences. Significant ($p < 0.05$) values are identified in bold, marginally significant ($p > 0.05$ and < 0.1) are identified in italics. Analytes marked with * have been $\log_{10}(x)$ transformed and analytes marked with ** have been $\log_{10}(x + 1)$ transformed. If “na”, then an ANOVA was conducted

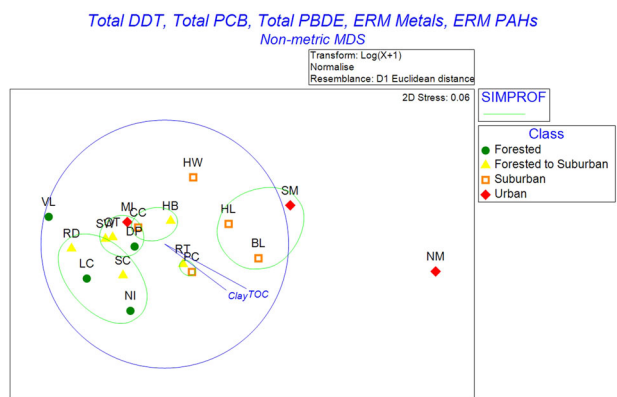


Fig. 5 Combined nMDS plots for total DDT, total PCB, total PBDE, and the metals and PAHs with ERM values. Oblong ovals indicate groupings of creeks based on similarity profile analysis. Clay and total organic carbon vectors are overlaid on the plot to identify their effect on creek distribution

Table 6 ANOSIM pair-wise test results comparing land-use classes

Pairwise comparison	R	p -value
Forested, Forested to Suburban	-0.10	0.7048
Forested, Suburban	0.53	0.0159
Forested, Urban	0.41	0.0571
Forested to Suburban, Suburban	0.30	0.0498
Forested to Suburban, Urban	0.43	0.0714
Suburban, Urban	0.21	0.1964

A p -value of 0.1 was used to determine statistical significance for this analysis. Significant values are identified in bold

Discussion

Concentrations of aluminum, arsenic, iron, manganese, and nickel are generally not enriched by anthropogenic sources, although it has been noted that high concentrations of arsenic in southeastern estuarine sediments may be related

to historical phosphate deposits (Windom et al. 1989). Unsurprisingly, however, their concentrations in this study were similar across watershed land-use classes. Concentrations of cadmium, chromium, copper, mercury, lead, and zinc, on the other hand, have been found to be increased by anthropogenic sources (Erlenkeuser et al. 1974; Goldberg et al. 1977; Sanger 1998). New Market Creek, an urban creek, had the highest concentration of each of these trace metals except for zinc, although concentrations of zinc in the forested creeks Crabhaul and Long were particularly high as well. Zinc particles from siding on buildings, automobile tire abrasion, fertilizer use, and natural deposits are sources of zinc in runoff (Agency for Toxic Substances and Disease Registry [ATSDR] 2005). Long Creek is surrounded by low-density housing but has a long history of agricultural activity. Crabhaul Creek has surrounding two-lane dirt roads that could contribute to tire-deposit runoff into the creek although not likely at significant levels. Therefore, it is unclear at this point what the sources of zinc are in these two forested systems.

Concentrations of cadmium, copper, mercury, and lead were found to be enriched primarily in suburban and urban creeks, although historical concentrations of these metals were similar to concentrations found in 2014 and 2015 (Sanger 1998; Sanger et al. 1999a). Outside of the Southeast, concentrations of these metals in New Market Creek, the most urbanized creek in this study, were similar to concentrations found in Elizabeth River, Virginia near a large naval installation (Conrad and Chisholm-Brause 2004). However, compared to known contaminated industrial sites in the Meadowlands of New Jersey (Artigas et al. 2017) concentrations in New Market Creek were much lower and were far more similar to the less-impacted suburban creeks in that study. Areas with high industrial or military activity often experience the same impacts to waterways as urban areas, due to the increased impervious cover, runoff, and chemical use.

Table 7 A summary of the paired analysis identifying whether creeks experienced an increase or decrease between 1994/1995 and 2014/2015

Creek name	Watershed land-use classification	Impervious cover	Clay	TOC	Mean ERM-Q	Mean Metal ERM-Q	Mean PAH ERM-Q	Total PAH	Total LMW PAH	Total HMW PAH	Total DDT	Total PCB
Deep	F	o	–	–	–	–	o	–	–	–	+	–
Crabhaul	F	o	–	+	+	+	o	+	–	+	o	–
Long	F	o	–	–	+	+	o	–	–	–	–	–
Sawmill	FS	+	–	–	–	–	o	+	–	–	–	o
Horlbeck	FS	+	–	–	–	–	+	+	+	+	–	o
Rathall	FS	+	–	o	–	–	o	–	–	+	+	+
Parrot	S	+	–	o	o	o	–	–	–	+	+	o
Cross	S	+	–	–	–	–	–	–	–	–	+	–
Bulls	S	+	–	+	+	o	+	+	+	+	+	+
Shem	U	+	–	+	+	+	+	+	–	+	+	–
New Market	U	+	–	+	+	+	+	+	+	+	+	+

The “o” indicates values stayed nearly the same over time, “+” indicates values increased, and “–” indicates values decreased. This table does not indicate significant changes

F Forested, FS Forested to Suburban, S Suburban, U Urban

Perylene and retene are known to naturally occur in the environment as they can be formed from early diagenesis (i.e., the alteration or breakdown) of plant pigments in the sediment, but they are also found anthropogenically in petroleum products, particularly perylene (Irwin et al. 1997). Retene was not related to the level of development; however, perylene was strongly related to IC, with concentrations significantly higher in the suburban and urban creeks than in the forested creeks, indicating the presence of an anthropogenic source of petroleum-based PAHs in these creeks. Nearly all other PAH concentrations were significantly higher in the urban creeks than in the forested creeks and were frequently higher than in the forested to suburban creeks. This makes sense, considering one of the most significant sources of PAHs is stormwater runoff with traces of PAHs from fossil fuel spills or leaks, and the combustion of fossil fuels (Hoffman et al. 1984; Eisler 1987; Irwin et al. 1997; Canadian Council of Ministers of the Environment [CCME] 1999).

LMW PAHs (e.g., naphthalene, acenaphthene, and fluorene) are acutely toxic, non-carcinogenic, and more volatile and water soluble relative to HMW PAHs, which allows for greater breakdown, dispersion, and bioaccumulation by aquatic organisms. They are generally petroleum-derived and from uncombusted fossil fuels. High molecular weight PAHs (e.g., fluoranthene, pyrene, and benzo(a)pyrene), however, are derived from the combustion of fossil fuels (Irwin et al. 1997) and have physio-chemical properties that allow them to adhere strongly to sediment particles and not easily degrade. They are generally not acutely toxic although they have been shown to have carcinogenic effects (Eisler 1987; CCME 1999). Crabhaul Creek was the only

creek with no LMW PAHs reported, but it had a total HMW PAH value greater than or similar to most of the forested to suburban creeks. The presence of HMW PAHs indicates the sources of PAHs in Crabhaul Creek were most likely combustion of gasoline in vehicles and/or atmospheric deposition from surrounding grass, wood, or coal burning.

In a study by Van Dolah et al. (2005), mean total PAH concentrations found in three tidal creeks in South Carolina near low use roads were similar to headwater concentrations in Deep Creek and Crabhaul Creek. Concentrations near moderate use roads were similar to concentrations in Hewlitts Creek, Bulls Creek, and Heyward Cove. However, in 2014 and 2015, concentrations in Shem Creek and New Market Creek far exceeded concentrations in any creeks sampled in Van Dolah et al. (2005), regardless of road traffic or distance from road. Similar concentrations between the two studies indicate that concentrations in TCP creeks are similar to those in other areas throughout South Carolina, and that Shem Creek and New Market Creek potentially have greater sources of PAHs relative to other typically developed areas, perhaps because of the considerably high level of boat traffic adjacent to and in these creeks.

Before being banned in 1972 due to adverse health effects to humans and wildlife (Simpson and Roger 1995) DDT was widely used in the United States for insect control on crops and in buildings. PCBs were used in electrical equipment, paint products, and industrial activities but were phased out of production and use by the US EPA in the 1970s. Both classes of contaminants are still found in the environment today due to their long half-lives, adherence to surface sediments, degradation into byproducts that also have

relatively long half-lives, and ability to bioaccumulate and biomagnify (National Pesticide Information Center [NPIC] 1999; ATSDR 2002; US EPA 2015). While no new DDT or PCBs are entering the environment, DDT byproducts are still being detected and PCBs could still be released from sources such as hazardous waste sites and leaks from old electrical equipment (US EPA 2015). In contrast, PBDEs are emerging contaminants of concern with their use starting in the 1970s as flame retardants in furniture, electrical equipment, and other household products. They attach to airborne particles and can be removed from the atmosphere via wet or dry deposition (ATSDR 2017). They also bind strongly to sediment particles, thus reducing their ability to breakdown quickly and leading to bioaccumulation, but environmental monitoring and concern has greatly reduced PBDE usage over the past decade (Wang et al. 2011; Noyes et al. 2013; ATSDR 2017). The highest concentrations of PCBs and PBDEs were observed in developed creeks, although total PCB seems to be decreasing.

The most abundant pesticides found in the creeks were 4,4'-DDD and 4,4'-DDE, both metabolites derived from the biodegradation of DDT. While total DDT was found to decrease over time, concentrations of 4,4'-DDE increased, which is expected as a breakdown product. Long Creek was historically surrounded by agricultural land which may explain why DDT residues were present in 2015. Total DDT in Long Creek, Hewlitts Creek, and New Market Creek were similar to or higher than total DDT levels observed in select urban creeks sampled in the Gulf of Mexico (Sanger et al. 2011). Furthermore, all suburban and urban creeks, and many forested and forested to suburban creeks, have current total DDT values higher than values observed in deeper Galveston Bay and Mississippi River Delta sites during peak DDT usage (1950s – 1960s) (Santschi et al. 2001). These comparisons further highlight how stressful headwater tidal creek habitats can be and present the ability of these systems to act as a long-term repository for pollutants.

Overall Impact

The US EPA (2012) considers sediment quality “poor” if there was at least one ERM exceedance, and “fair” if no ERMs were exceeded but five or more ERLs were exceeded. Using these guidelines, the creeks sampled during 2014/2015 with the highest potential for biological effects were the urban creeks Shem Creek and New Market Creek. Based on US EPA thresholds and the definition by Long et al. (1995), sediment quality in both Shem Creek and New Market Creek would be considered “poor” and it would seem likely that concentrations in these contaminated sediments were high enough to impact benthic communities. Among the other developed creeks, Heyward Cove,

Rose Dhu, and Stoney would be considered “poor”, as well as the forested creeks Crabhaul and Long. Additionally, each of these creeks (except for Rose Dhu), as well as Bulls and Hewlitts, had mean ERM-Q values >0.058. Hyland et al. (1999) identified mean ERM-Q values >0.058 as being high-risk and potentially indicative of degraded habitat and benthic community assemblages. Impacts to the benthic community include observed loss of species richness and the dominance of few select opportunistic species (Parker 2018). Macro-benthic invertebrates are a link between food sources and predators, and a community dominated by only a few species could eliminate target prey species for organisms at higher trophic levels.

An underlying focus of this study was to consider a new class of creeks, with forested watersheds in the early 1990s, that were developed and categorized as suburban by 2016. Horlbeck, Okatee, and Sawmill creeks would have been reclassified around 1996, Rathall Creek around 2001, Rose Dhu Creek around 2006, and Stoney Creek just recently around 2016 (McHouell 2016). Increased development has been associated with impact to sediment quality and over time these TCP creeks support this trend, but responsible development may help mitigate these impacts. Several tools or BMPs are now used in coastal development in order to decrease and protect against potential impacts. Stormwater ponds are one of the many BMPs implemented in modern developments but BMPs also include vegetative buffers, minimization of watershed IC, and using pervious surface alternatives. BMPs are meant to slow the rate of stormwater runoff and aid in filtering particulates and contaminants before they reach nearby waterways (Muthukrishnan et al. 2005). Weinstein et al. (2010) and Crawford et al. (2010) identified levels of sediment contamination in stormwater ponds in coastal South Carolina. They found that ponds surrounded by commercial development accumulate significantly high levels of metals, PAHS, and some pesticides, indicating that they successfully sequester contaminants and will presumably decrease the contaminants received by nearby tidal creeks.

Among the forested to suburban study creeks, Stoney Creek most recently experienced watershed development around 2016, with the number of coastal manmade ponds in this small watershed increasing from 16 to 22 between 2006 and 2013 (McHouell 2016), yet contaminant levels were still similar to forested creeks. It is possible that the implementation of these BMPs during development slowed the accumulation of contaminants in the creek, but it may also be that contamination simply lags behind development and that at the time of sampling contaminant levels in Stoney Creek had not increased significantly. On the other hand, Rathall Creek experienced a significant increase in percent IC in its watershed, jumping from 10% in 2001 to 43% in 2016, yet only four stormwater ponds were constructed, around 2006

which could partially account for higher levels of contaminants reaching the creek which caused concentrations to be similar to historically suburban creeks. Interestingly, Rose Dhu and Okatee creeks have been developed for a number of years, yet contamination is similar to forested creeks rather than suburban creeks. This may be because both creeks experienced gradual increases in watershed IC and had high numbers of stormwater ponds during development. Okatee Creek has a large watershed size of 2,411 hectares, but between 2006 and 2013 the number of stormwater ponds increased from 84 to 124. Rose Dhu Creek already had 30 ponds in place in 2006, around when its primary increase in watershed IC occurred (McHouell 2016).

Summary

Overall, the suburban and urban creeks have greater contaminant levels than the forested and forested to suburban creeks. In many cases, current relationships between contaminant metrics, individual analytes, and watershed IC were similar to historical relationships, thus reaffirming long-term relationships identified between coastal development and sediment contamination, as well as indicating a continued introduction of chemicals into the environment or the impact of long half-lives. Forested to suburban creek contamination values frequently fell above forested values but below suburban values, indicating an increase in pollutants over time but not enough to reach levels similar to historically suburban creeks. This could be because accumulation is still occurring or because BMPs have been playing their intended role in slowing the rate of pollution influx into the creeks. Regardless, it seems likely that preventing excess contamination in tidal creeks surrounded by development is reliant on not only implementing the use of BMPs before and during any new construction, but also managing development to occur at a slower rate.

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