

# Landscape and Watershed Processes

## Landscape Metrics and Estuarine Sediment Contamination in the Mid-Atlantic and Southern New England Regions

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### ABSTRACT

In a previously published study, quantitative relationships were developed between landscape metrics and sediment contamination for 25 small estuarine systems within Chesapeake Bay. These analyses have been extended to include 75 small estuarine systems across the mid-Atlantic and southern New England regions of the USA. Because of the different characteristics and dynamics of the estuaries across these regions, adjustment for differing hydrology, sediment characteristics, and sediment origins were included in the analysis. Multiple linear regression with stepwise selection was used to develop statistical models for sediment metals, organics, and total polycyclic aromatic hydrocarbons (PAHs). The landscape metrics important for explaining the variation in sediment metals levels ( $R^2 = 0.72$ ) were the percent area of nonforested wetlands (negative contribution), percent area of urban land, and point source effluent volume and metals input (positive contributions). The metrics important for sediment organics levels ( $R^2 = 0.5$ ) and total PAHs ( $R^2 = 0.46$ ) were percent area of urban land (positive contribution) and percent area of nonforested wetlands (negative contribution). These models included silt-clay content (metals) or total organic C (organics, total PAHs) of sediments and grouping by estuarine hydrology, suggesting the importance of sediment characteristics and hydrology in mitigating the influence of the landscape metrics on sediment contamination levels. The overall results from this study are indicative of how statistical models can be developed relating landscape metrics to estuarine sediment contamination for distributions of land cover and point source discharges.

THE EMPHASIS ON CONTROL OF pollution sources over the last decade in the USA has shifted from point to nonpoint sources, the result of Congressional amendments to the Clean Water Act in 1987 (Navigation and Navigable Waters Act, 1994; USEPA, 1991). Runoff is the major source of nonpoint pollution of surface water, with the magnitude of the polluted runoff dependent upon the land cover, land use, and management practices employed (Basnyat et al., 1999). The interest in pollution control is for the protection and maintenance of aquatic resources within proximity of these land areas. A recent National Water Quality Inventory (USEPA, 1995) indicated that runoff from urban areas was the largest source of water quality impairments to estuaries.

From several perspectives, the development of relationships between point sources of pollution and condition of aquatic resources is relatively straightforward compared with the development of comparable relationships for nonpoint sources. First of all, by definition, nonpoint pollution is more diffuse, and thus harder to

quantify. It is widespread over the land surface, for example, by atmospheric deposition or application of agricultural pesticides (Patty et al., 1999; Mason et al., 1999; Sherrell and Ross, 1999). Secondly, the land, and the cover on it, acts as a filter, retarding the overland flow of pollutants, diffusing them even further, and providing a medium for geochemical transformations (Correll et al., 1992; Schueler, 1994; Lajtha et al., 1995; Munn and Gruber, 1997; Wahl et al., 1997; Thierfelder, 1998). Finally, the pollution can be directed downward into the ground water, providing for even more complex pathways (Hughes et al., 1998; Hussain et al., 1999; Li et al., 2000).

Measures of the characteristics of land cover, land use, and management practices have been of significant interest in recent literature (O'Neill et al., 1988; McGarigal and Marks, 1995; Wickham and Riitters, 1995; Jones et al., 1996, 1997; Riitters et al., 1995; Thierfelder, 1998; O'Neill et al., 1999). As a result, many different landscape metrics have been developed. A relevant question is how does one determine "which, if any, of these landscape metrics are appropriate for predicting the impact of nonpoint pollution on estuarine receiving waters?"

Comeleo et al. (1996) reported a study of 25 small estuarine systems in Chesapeake Bay to assess possible relationships between estuarine sediment contaminant levels and landscape metrics in the watershed. The area of developed land located in the watershed within 10 km of the sediment sampling station was a major contributing factor in the sediment concentrations of both metals and organic contaminants. Limitations to this pilot study included the absence of a wetlands class for the land cover data set used for development of the landscape metrics (USEPA, 1994), the limited number of watersheds (25), and the unknown transferability of results beyond the relatively low-relief, coastal plain watersheds investigated. The purposes of the study reported in this paper were to (i) address these limitations so that the resulting conclusions for estuarine sediment contamination would be applicable to other geographic areas and (ii) develop approaches that would have predictive capabilities, for use with other data sets.

**Abbreviations:** ANOVA, analysis of variance; CV, coefficient of variation; DEM, digital elevation model; EMAP, Environmental Monitoring and Assessment Program; GIS, geographic information system; LUDA, land use data analysis; MLRSS, multiple linear regression with stepwise selection; NOAA, National Oceanic and Atmospheric Administration; PAH, polycyclic aromatic hydrocarbon; PC1, first principal component; PC2, second principal component; PCA, principal component analysis; PCB, polychlorinated biphenyl; TOC, total organic carbon; USEPA, U.S. Environmental Protection Agency; USGS, U.S. Geological Survey; VIF, variance inflation factor.

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## METHODS

### Study Area

Small estuarine systems (<260 km<sup>2</sup> in estuarine surface area) in the Virginian Biogeographic Province (Cape Cod to mouth of Chesapeake Bay on the east coast of the USA; Holland, 1990) and their associated watersheds were used for the study (Fig. 1). This province covers the mid-Atlantic and southern New England regions of the USA. Small estuarine systems were chosen because their physical characteristics

(small in areal extent, semi-enclosed, relatively shallow, in close proximity to land-based stressors) enhance the possibility of coupling sediment condition to surrounding watershed conditions. In analysis of benthic community data collected in the summers of 1990–1993 across the estuarine waters of the Virginian Biogeographic Province, the primary stressor influencing benthic community conditions in small estuarine systems was found to be sediment contamination (Paul et al., 1999). The number of estuarine systems was 75 out of a possible 144 systems because of available quality-assured sediment

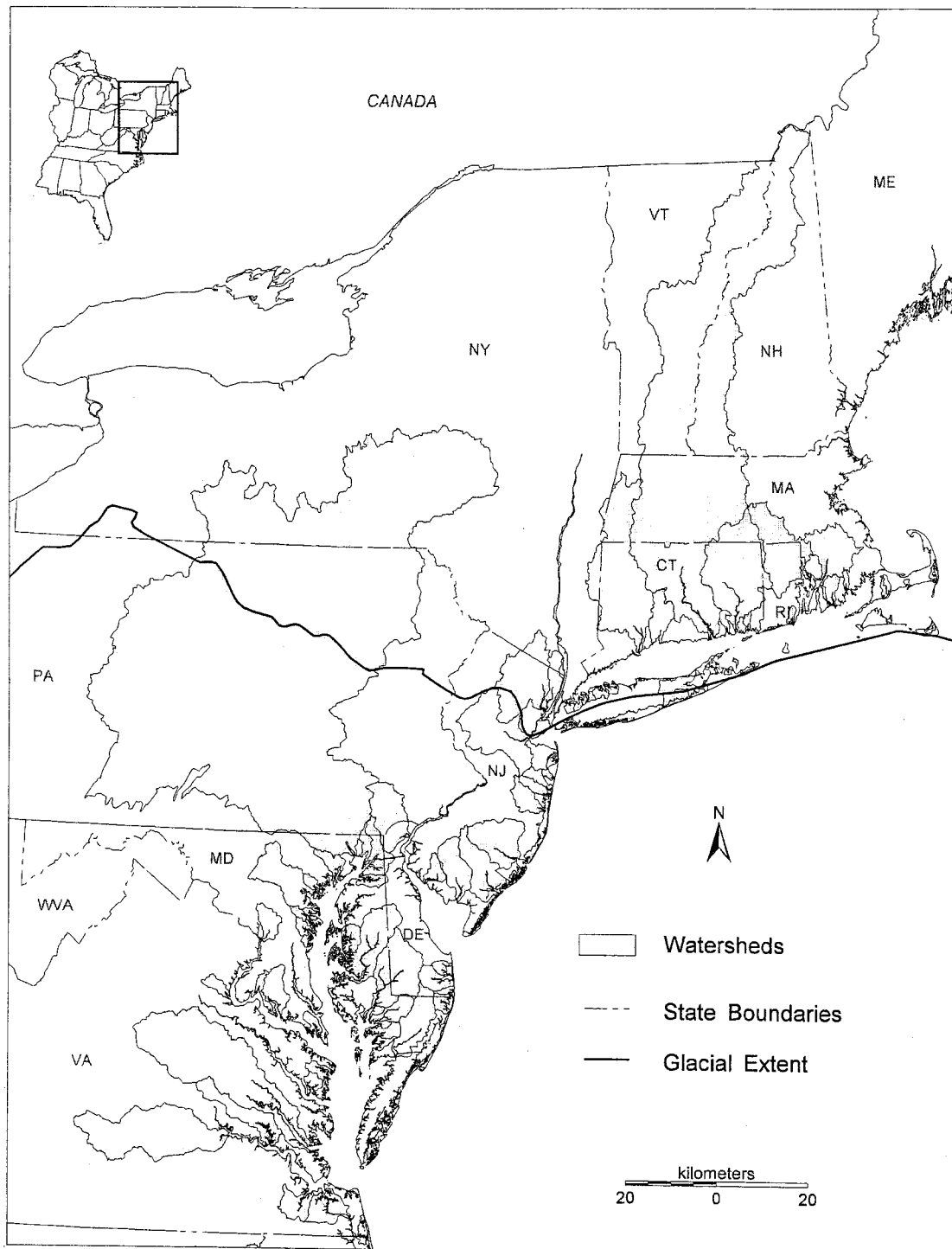


Fig. 1. Distribution of estuarine watersheds in the mid-Atlantic and southern New England regions used in this study.

contaminant data for both metals and organics and due to the inability to accurately delineate watersheds on some of the relatively flat coastal areas (see below under Watershed Delineation). The 75 watersheds are distributed throughout the Province (Fig. 1). These included the 25 systems in Chesapeake Bay used in Comeleo et al. (1996). Thirty-two were in Chesapeake Bay, with the rest distributed northward to Cape Cod.

### Data Sources

Sediment contaminant data were collected in the small estuarine systems of the Virginian Province during the summers of 1990–1993. One hundred forty-four small estuarine systems (<260 km<sup>2</sup> in estuarine surface area) were selected for sampling, with probability survey designs used to sample all of the estuarine waters in the province (Holland, 1990; Strobel et al., 1995). For sample collection within each small system, a single sampling site was randomly selected. The top 2 cm of sediments were composited from multiple samples collected from each site with a grab sampler (440 cm<sup>2</sup> surface area) and subsequently analyzed for a suite of organic and inorganic constituents. Sediment samples were analyzed for 24 polycyclic aromatic hydrocarbons (PAHs), 18 polychlorinated biphenyl (PCB) congeners, DDTs, 11 chlorinated pesticides, tributyl tins, and 15 metals (Strobel et al., 1995, 1999). Of these, 9 metals and 13 organic compounds shown to cause measurable effects on benthic organisms (Long et al., 1995) were selected for further consideration as dependent variables (Table 1). In addition, total PAHs, the combination of the 24 measured individual PAHs (NOAA, 1991), was used in the analyses. Nondetectable values were assigned zeros.

Two landscape metric categories were used for analysis: land cover pattern and point source pollution input. Land cover pattern, for this study, refers to the composition of land cover types within a watershed. We assumed that land cover pattern is a suitable surrogate for nonpoint-source pollution information, as it has been shown that the severity of nonpoint-source pollution is closely related to land cover (Kim and Ventura, 1993). Land cover data were from the U.S. Geological Survey Land Use Data Analysis (LUDA) data set (Fegeas et al., 1983), which was interpreted from 1970s aerial photos. This data set was the only available consistent land cover classification for the entire Virginian Province estuarine watershed at the initiation of this study. The 37 LUDA land cover classes were aggregated to be compatible with the land cover classes used in Comeleo et al. (1996): urban (comparable to developed land), forested, agricultural, forested wetlands, and nonforested wetlands (comparable with herbaceous land).

Point source pollution data were obtained from the NOAA National Coastal Pollutant Discharge Inventory for the estuarine drainage areas of the Virginian Province (Pacheco, 1993). The location of active major point source discharge sites as

well as 1991 estimates of annual pollutant input (kg/yr) and annual outflow (10<sup>6</sup> m<sup>3</sup>/h) at each discharge site were used for analysis. Estimates were made from effluent monitoring data available in monthly Discharge Monitoring Reports completed as part of each facility's National Pollutant Discharge Elimination System Compliance Monitoring Program (Pacheco, 1993). Effluent data were available for seven (As, Cd, Cr, Cu, Pb, Hg, and Zn) of the nine metals in Table 1. These seven metals are present in the environment from natural and anthropogenic sources and are among the most frequently measured pollutants at monitored discharge sites (Pacheco, 1993). Effluent data for organics were sparse and were not considered for further analysis.

Response of estuarine systems to the watershed stressors is expected to be mitigated by the complex dynamics occurring within the individual systems. Three ways to account for the dynamics were explored: (i) including grain size and total organic C content of bottom sediments as possible variables in the statistical models; (ii) grouping systems by whether or not they were glaciated; and (iii) grouping systems on the basis of estuarine hydrology.

Sediment grain size (SICL) and total organic carbon (TOC) were separately included as possible independent variables to account for sediment characteristics in the statistical models. Grain size, expressed as percent silt-clay content, and TOC (percent of sediment) were analyzed on samples collected with those for contaminant analyses. Silt-clay content and TOC are indicative of the sedimentary environment of the system and result from the interaction of loadings, circulation, and geomorphology (Nichols and Biggs, 1985). Total organic C is also indicative of biological processes (eutrophication) in the system. Contaminants tend to partition preferentially on fine grain particles, particularly onto the organic fraction (Landrum and Robbins 1990; Bierman, 1994).

Estuarine sediments north of the southern extent of the Wisconsin glaciation are composed of stratified proglacial deposits of outwash plains (Golet et al., 1993). South of the glaciated area, sediments are the result of continental surface erosion, transport, and deposition. The transport and fate of particle-bound contaminants can be influenced by the origin of the sediments (Bonner et al., 1994). A categorical variable (GLACIAL) was used to separate glacial ( $n = 30$ ) from nonglacial ( $n = 46$ ) estuarine systems.

Hydrology was represented by the ratio of estimated freshwater inflow to estimated tidal volume (Kennish, 1986). U.S. Geological Survey stream flow records were used to quantify inflows, with ungaged flow estimated using watershed area factors. We defined hydrology classes as follows for our analyses. Inflow was greater than tidal volume in Class 1 estuaries, approximately equal to tidal volume for Class 2 estuaries, and less than tidal volume for Class 3 estuaries. Class 4 estuaries were shallow, heavily influenced by surface processes, and included barrier beach/lagoonal systems. The estuarine circulation dynamics control the transport and ultimate dilution of material entering the systems (Fischer et al., 1979). A categorical variable (HYDROLOGY) was used to separate Class 1/Class 2 estuarine systems ( $n = 51$ ) from Class 3/Class 4 systems ( $n = 24$ ).

### Watershed Delineation

A watershed approach (Comeleo et al., 1996) was used to analyze the landscape surrounding the sampling site in each estuary. Watershed delineation was attempted for each small estuarine system using one-degree (1:250 000 scale) U.S. Geological Survey (USGS) Digital Elevation Model (DEM) data processed with the GRID module of the Environmental Sys-

**Table 1. Sediment contaminants used in analyses as possible dependent variables.**

Metals	Organics
Silver	Total PAHs†
Arsenic	Acenaphthene
Cadmium	Acenaphthylene
Chromium	Anthracene
Copper	Fluorene
Mercury	2-Methylnaphthalene
Nickel	Naphthalene
Lead	Phenanthrene
Zinc	Benz(a)anthracene
	Benzo(a)pyrene
	Chrysene
	Dibenz(a,h)anthracene
	Fluoranthene
	Pyrene

† Combination of 24 measured individual PAH compounds (NOAA, 1991).

tem Research Institute (ESRI) ARC/INFO GIS software package. The accuracy of computer delineation for many of the low relief Coastal Plain watersheds was questionable because the horizontal and vertical resolution of the elevation data (93 and 1 m, respectively) were inadequate to discern low topographic relief. The computer-derived watersheds were reviewed and enhanced through on-screen interpretation using DEM-derived shaded relief maps and hydrography (1:100 000 USGS Digital Line Graph) as backdrops. In areas of low topographic relief where the accuracy of computer-interpreted watershed boundaries was questionable, divisions between drainage systems were manually located by interpolation. The delineated watersheds were combined with available sediment contaminant data. This resulted in 75 nonnested estuarine/watershed systems for analysis.

**Landscape Analysis**

The vector module of ARC/INFO GIS software was used to determine the area of each land cover class found within each watershed and the total annual outflow and metals input from all point discharge sites located within each watershed. A total of 14 landscape metrics were calculated: area of watershed (AREA), percent area of urban land (PURB), percent area of agricultural land (PAG), percent area of forested land (PFOR), percent area of forested wetlands (PFWET), percent area of nonforested wetlands (PNFWET), and the annual volume (FLOW) and metals input (LAS, LCD, LCR, LCU, LPB, LHG, LZN) from point discharge sites.

**Statistical Analysis**

Principal component analysis (PCA) was used to reduce the number of variables for statistical modeling. Principal component analysis was performed on the correlation matrix of the sediment contaminant and point source input variables. Principal component analysis was performed separately on the group of nine metals and the group of 13 organic compounds in the sediments. Principal component analysis also was performed on the seven metals input rates.

Pairwise correlations were calculated as measures of association between each of the independent variables and the dependent variables used to develop the statistical models. Multiple linear regression with stepwise selection (MLRSS, a.k.a. stepwise MLR) was conducted to develop statistical models between the dependent variables (the first principal component for sediment metals and organics, and total PAHs) and independent variables (SICL, TOC, GLACIAL, HYDROLOGY, and landscape metrics: AREA, PURB, PAG, PFOR, PFWET, PNFWET, FLOW, and the first principal component for annual metals input). Refer to Table 2 for summary of

the independent variables. GLACIAL was a categorical variable (0/1) to separate glacial from nonglacial systems, respectively. HYDROLOGY was a categorical variable (0/1) to separate Class 1 and 2 from Class 3 and 4 systems, respectively. A significance level of  $p < 0.15$  was used for independent variables to enter the model. Two basic forms of statistical models were investigated with MLRSS:

$$y = \alpha + \sum \beta_i x_i + \epsilon, \text{ and} \tag{1}$$

$$\ln(y + \text{offset}) = \alpha + \sum \beta_i x_i + \epsilon \tag{2}$$

where  $y$  is the dependent variable,  $x_i$  are independent variables,  $\alpha$  and  $\beta_i$  are regression coefficients, offset is a constant, and  $\epsilon$  is the residual error. An offset was used in Eq. [2] with natural log transform of the dependent variable to accommodate zeros (total PAHs) and negative values (principal components).

Potential models relating landscape metrics to sediment contamination were investigated with MLRSS for the following combinations: three dependent variables (PC1-metals, PC1-organics, total PAHs), two functional forms [linear and exponential (a.k.a. log transform of dependent variable)], and seven possible combinations of independent variables. The independent variable combinations are indicated in Table 2. Combination A treats the two categorical variables as potential additive variables in the regression (i.e., additional  $x_i$  in Eq. [2] with values of 0 or 1), resulting in a single-equation model. For this model, the coefficients for each of the independent variables are not a function of the categorical variables. Combinations B and C exclude variables for sediment origin (GLACIAL) and estuarine hydrology (HYDROLOGY) from the regression, and result in single-equation models. For combinations D through G, two-equation models (or nested regressions) were developed, with a different equation for each value of the categorical variable GLACIAL (D and F) and HYDROLOGY (E and G). These combinations treat GLACIAL and HYDROLOGY as grouping variables (not as additional  $x_i$  in Eq. [2]), that is, the coefficients for the independent variables are functions of the categorical variable [for example,  $\alpha = \text{function}(\text{GLACIAL})$  and  $\beta_i = \text{function}(\text{GLACIAL})$ ].

Each fitted model ( $3 \times 2 \times 7$  possible combinations) was tested for validity of regression assumptions by examining the distribution of residuals (Q-Q normal probability plots and Shapiro-Wilk test for normality). Variance inflation factors (VIFs) were calculated to identify correlations among independent variables in the fitted models. Variance inflation factor is a measure of the amount that the variance in a regression coefficient is inflated due to multicollinearity of variables. A VIF greater than 10 for a variable indicates multicollinearity

**Table 2. Combinations of independent variables considered in regression analysis. Abbreviations for variables in parentheses.**

Independent variable	Independent variable combinations†						
	A	B	C	D	E	F	G
Estuarine hydrology (HYDROLOGY)	X‡				C§		C§
Glaciated sediments (GLACIAL)	X‡			C§		C§	C§
Sediment grain size (SICL)	X	X		X	X		
Sediment total organic carbon (TOC)	X		X			X	X
Area of watershed (AREA)	X	X	X	X	X	X	X
Percent area of urban land (PURB)	X	X	X	X	X	X	X
Percent area of agricultural land (PAG)	X	X	X	X	X	X	X
Percent area of forested land (PFOR)	X	X	X	X	X	X	X
Percent area of forested wetlands (PFWET)	X	X	X	X	X	X	X
Percent area of nonforested wetlands (PNFWET)	X	X	X	X	X	X	X
Point source annual outflow (FLOW)	X	X	X	X	X	X	X
Point source metals input (PCI-INPUT)	X	X	X	X	X	X	X

† X, considered as additive independent variable. C, treated as categorical (grouping) variable.  
 ‡ As additive variable, value of 0 or 1.  
 § As grouping variable, regression coefficients for additive variables are function of this variable.

**Table 3. Summary statistics for sediment contaminants for systems ( $n = 75$ ) used in analyses.**

Sediment contaminant	Mean	SD†	CV‡	Median	Minimum§	Maximum
mg/kg						
<b>Metals</b>						
Silver	0.8	1.6	205.5	0.1	0.0	9.7
Arsenic	9.8	7.1	72.5	8.8	0.7	36.2
Cadmium	0.8	1.2	140.5	0.4	0.0	6.6
Chromium	71.6	67.4	94.2	57.8	2.4	348.0
Copper	66.2	104.3	157.5	28.5	1.1	680.0
Mercury	0.3	0.4	150.9	0.1	0.0	2.0
Nickel	24.9	21.1	84.8	19.8	0.0	136.0
Lead	60.7	62.5	103.0	40.5	0.0	323.0
Zinc	169.2	147.8	87.4	132.0	5.0	797.0
µg/kg						
<b>Organics</b>						
Acenaphthene	113	622	550	3	0	5 120
Acenaphthylene	24	95	405	0	0	803
Anthracene	186	759	409	15	0	6 010
Fluorene	89	306	345	15	0	2 320
2-methylnaphthalene	73	120	164	20	0	489
Naphthalene	103	188	182	23	0	1 120
Phenanthrene	534	1 991	373	73	0	12 800
Benz(a)anthracene	627	3 397	542	57	0	29 100
Benzo(a)pyrene	307	855	279	57	0	6 010
Chrysene	549	2 272	414	78	0	18 400
Dibenz(a,h)anthracene	114	664	584	6	0	5 740
Fluoranthene	1 164	5 345	459	112	0	42 200
Pyrene	994	4 042	407	137	0	31 800
Total PAHs	7 223	29 293	406	1 069	0	240 620
%						
Sediment grain size (silt and clay)	59.2	33.7	57.0	71.7	0.6	99.4
Total organic carbon (TOC)	2.3	1.3	55.9	2.4	0.1	7.0

† Standard deviation.

‡ Coefficient of variation, %.

§ Nondetectable values were assigned zero.

(Philippi, 1993), and an ambiguity in the interpretation of regression coefficients. For each dependent variable and functional form, the best overall model was determined by the coefficient of determination ( $R^2$ ) of the fitted models. The best overall model was compared with the other fitted models for the same dependent variable and functional form by testing for significant difference (ANOVA with  $p < 0.05$ ). Statistical analyses were done using SAS (SAS Inst., 1989) and S-Plus 2000 (MathSoft, 1999) software.

## RESULTS

### Sediment Contaminants

The characteristics of the sediment contaminant data for the 75 systems are presented in Table 3. The sediment metals were less variable across the systems compared with the sediment organics based on the coefficient of variation (CV). Two organic compounds (2-methylnaphthalene and naphthalene) had lower variability than the other compounds. These two PAHs have the lowest molecular weights. For total PAHs and the individual compounds used in the analysis, very strong correlations exist except for three compounds (fluorene, 2-methylnaphthalene, and naphthalene), two of which had lower variability across systems (CV in Table 3). Total PAHs was strongly correlated with the first principal component for the 13 organic compounds but not correlated with the second principal component. So PC1-ORG can be interpreted as a good measure for total PAHs.

### Landscape Characteristics

The characteristics of the 75 watersheds used in the analysis varied widely (Table 4). Watersheds ranged in

size from 6 to 70 800 km<sup>2</sup>. The percentage of urban land in watersheds ranged from 2 to 99% and was the most abundant land cover in 17 of the 75 watersheds. Forested land was the most abundant land cover in 43 and agricultural land in 15 watersheds. Point source outflow and metals input also varied widely among watersheds. The characterization of systems for glacial-nonglacial systems and Class 1 and 2/Class 3 and 4 estuaries are also presented in Table 4. Comparing mean values, the Class 3 and 4 system watersheds were much smaller in area, were more urbanized, had less agricultural lands, and had lower point source input. These watersheds received much less metals loadings, with chromium input being the exception. The glaciated watersheds were smaller, more urbanized, had less agricultural lands, and had higher point source flows and metals input.

### Statistical Results

The first principal component for each of the sediment metals and organics showed strong correlations between principal component and sediment concentration for nearly all contaminants in the 75 systems (Table 5). Arsenic, which was weakly correlated with PC1, was the only metal strongly correlated with PC2. The three organic compounds weakly correlated with total PAHs (fluorene, 2-methylnaphthalene, and naphthalene) were weakly correlated with PC1, but the two with low variability (low CV) were strongly correlated with PC2 (negatively). We interpret PC1 to reflect the overall amount of metals or organic contaminants: the larger PC1, the greater the magnitude of sediment contamination. The

**Table 4. Summary statistics for landscape metrics for grouping of systems (Virginian Province, glaciated/nonglaciated, and hydrology class) used in analyses.**

Landscape metric	All Virginian Province watersheds (n = 75)						Virginian Province glaciated watersheds (n = 30)						Virginian Province nonglaciated watersheds (n = 45)					
	Mean	SD	CV‡	Median	Min	Max	Mean	SD	CV‡	Median	Min	Max	Mean	SD	CV‡	Median	Min	Max
Area, km <sup>2</sup>	2 022	8 743	432	224	6	70 818	1 557	5 286	340	72	6	28 807	2 332	10 484	450	413	60	70 818
Urban land, %	26	25	96	16	2	99	38	29	77	24	6	99	18	19 101	13	2	78	
Agricultural land, %	21	19	92	13	0	74	7	7 106	5	0	32	31	19	62	28	3	74	
Forested land, %	43	22	51	44	0	78	44	28	65	48	0	78	43	17	40	43	10	74
Forested wetlands, %	2	5	215	0	0	21	1	2 193	0	0	9	3	6	195	0	0	21	
Nonforested wetlands, %	2	5	201	1	0	27	1	3 186	0	0	9	3	6	188	1	0	27	
Annual outflow, 10 <sup>6</sup> m <sup>3</sup> /yr	850	3 957	466	2	0	32 795	1 968	6 139	312	0	0	32 795	105	281	267	2	0	1 397
Arsenic input, kg/yr	1 876	4 849	258	3	0	30 814	3 076	6 710	218	0	0	30 814	1 076	2 870	267	27	0	14 530
Cadmium input, kg/yr	826	2 163	262	2	0	13 548	1 341	3 000	224	0	0	13 548	483	1 280	265	14	0	5 642
Chromium input, kg/yr	5 105	17 713	347	32	0	129 349	10 260	27 001	263	0	0	129 349	1 669	4 122	247	98	0	20 709
Copper input, kg/yr	5 024	19 101	380	55	0	149 285	10 330	29 331	284	0	0	149 285	1 487	3 818	257	81	0	16 836
Lead input, kg/yr	2 975	8 396	282	0	0	58 711	5 178	12 104	234	0	0	58 711	1 506	4 058	269	30	0	20 732
Mercury input, kg/yr	23	63	272	0	0	446	41	92	222	0	0	446	11	28	250	1	0	137
Zinc input, kg/yr	23 309	100 032	429	0	0	827 952	48 839	154 948	317	0	0	827 952	6 289	16 256	258	314	0	78 736
	Virginian Province Class 1 and 2 watersheds (n = 51)						Virginian Province Class 3 and 4 watersheds (n = 24)											
Area, km <sup>2</sup>	2 914	10 516	361	467	59	70 818	127	160	126	69	6	655						
Urban land, %	21	23	105	13	2	99	37	28	76	24	10	98						
Agricultural land, %	26	19	73	25	0	74	10	13	140	5	0	47						
Forested land, %	45	19	44	46	0	78	40	27	67	41	0	77						
Forested wetlands, %	2	5	198	0	0	21	1	4	275	0	0	17						
Nonforested wetlands, %	3	5	210	1	0	27	2	3	159	1	0	9						
Annual outflow, 10 <sup>6</sup> m <sup>3</sup> /yr	1 122	4 743	423	3	0	32 795	271	981	362	0	0	4 779						
Arsenic input, kg/yr	2 285	5 386	236	61	0	30 814	1 008	3 381	336	0	0	14 150						
Cadmium input, kg/yr	1 039	2 464	237	22	0	13 548	375	1 243	332	0	0	4 950						
Chromium input, kg/yr	4 585	11 846	258	129	0	64 359	6 210	26 506	427	0	0	129 349						
Copper input, kg/yr	6 823	22 854	335	96	0	149 285	1 203	4 001	333	0	0	16 297						
Lead input, kg/yr	3 786	9 669	255	85	0	58 711	1 251	4 358	348	0	0	19 582						
Mercury input, kg/yr	30	73	247	1	0	446	10	32	327	0	0	131						
Zinc input, kg/yr	32 060	120 181	375	498	0	827 952	4 714	16 276	345	0	0	72 327						

† Standard deviation.

‡ Coefficient of variation, %.

first principal components for metals (PC1-MET) and organics (PC1-ORG) were used to represent the level of sediment contamination in the regression analyses in addition to total PAHs, with 63 and 83% of standardized variance explained, respectively.

The result of PCA with metals input variables indicated strong correlations for all loading variables in PC1 except Zn, which was the only loading in PC2 (Table 5). The PC1-INPUT explained 68% of the standardized variance in metals input. The PC1-INPUT was used to represent the metals input in the regression analyses.

The pairwise correlations between PC1-MET, PC1-ORG, and total PAHs and the independent variables can be summarized as follows. Sediment metals (PC1-MET) significantly correlated with sediment characteristics (SICL and TOC), nonpoint sources (positive for PURB and negative for PFOR and PNFWET), and point sources (FLOW and PC1-INPUT). Sediment organics (PC1-ORG) was correlated with sediment characteristics (primarily TOC) and nonpoint sources (positive for PURB and negative for PFOR). Total PAH correlations were similar to organics except PAG was substituted for PFOR. In almost all cases, urban land exhibited the strongest correlation with sediment contamination.

The multiple linear regression with stepwise selection results for statistical models with Eq. [2] were consistently found to have normally distributed residuals. The residuals for all models with Eq. [1] were nonnormal. These results are summarized in Table 6. Except for

one case, the VIFs for the fitted models were much less than 10, indicating little concern for multicollinearity of variables. The exception exhibited strong correlation ( $|r| > 0.9$ ) among PURB, PAG, and PFOR for nonglacial systems. In addition to the  $R^2$  for each model and dependent variable, those models shown to be not statistically different from that with largest  $R^2$  (ANOVA with  $p \leq 0.05$ ) are indicated. A summary of these latter models is shown in Table 7 with coefficients that entered the regression with  $p \leq 0.15$ .

## DISCUSSION

The results of this study have shown a quantitative linkage between land use and sediment contamination for small estuarine systems. The sediment contaminant levels in small estuarine systems increase with increasing percent of the watershed in urban land. This is consistent with the conclusion of a recent National Water Quality Inventory (USEPA, 1995) that runoff from urban area is the prime source of water quality impairments in estuaries. Runoff from urbanized areas is a major source of nonpoint pollution (Wang et al., 1997). The amount of impervious surface resulting from urbanization influences the condition of receiving waters (Boward et al., 1999). Dauer et al. (2000) observed strong correlations between sediment contamination and percent urban area in the watersheds of 10 major tributaries of Chesapeake Bay.

Sediment contaminant levels decrease with increasing

**Table 5. Loadings (for absolute values > 0.1) and correlation coefficients (*r*) for the first (PC1) and second (PC2) principal components for sediment metals, sediment organic contaminants, and metals input from point sources with individual contaminant concentrations and metal loadings.**

Variable	PC1	<i>r</i>	PC2	<i>r</i>
<b>Sediment metals</b>				
Silver	0.323	0.77	-0.427	-0.48
Arsenic	0.179	0.43	0.628	0.71
Cadmium	0.379	0.9	-0.172	-0.19
Chromium	0.362	0.86	0.202	0.23
Copper	0.333	0.79		
Mercury	0.329	0.78	-0.42	-0.47
Nickel	0.311	0.74	0.334	0.38
Lead	0.355	0.84		
Zinc	0.383	0.91	0.237	0.27
% Variation explained	63%		14%	
<b>Sediment organics</b>				
Acenaphthene	0.298	0.98	0.166	0.19
Acenaphthylene	0.286	0.94		
Anthracene	0.302	0.99		
Fluorene	0.236	0.78	-0.144	-0.16
2-methylnaphthalene	0.140	0.46	-0.780	-0.88
Naphthalene	0.247	0.81	-0.467	-0.53
Phenanthrene	0.283	0.93		
Benz(a)anthracene	0.292	0.96	0.178	0.20
Benz(a)pyrene	0.287	0.94	-0.116	-0.13
Chrysene	0.302	0.99		
Dibenz(a,h)anthracene	0.285	0.94	0.196	0.22
Fluoranthene	0.300	0.98	0.127	0.14
Pyrene	0.302	0.99		
% Variation explained	83%		10%	
<b>Point source metals input</b>				
Arsenic input	0.447	0.98		
Cadmium input	0.437	0.95		
Chromium input	0.329	0.72		
Copper input	0.251	0.55	-0.322	-0.30
Lead input	0.446	0.97		
Mercury input	0.449	0.98		
Zinc input	0.193	0.42	0.933	0.88
% Variation	68%		13%	

percent of the watershed in nonforested wetlands. These wetlands are generally located in the lower parts of the watershed, closer to the shoreline, and function as a buffer between the urban areas and receiving waters. Herbaceous land within 10 km of the sampling site was a surrogate for nonforested wetlands in Comeleo et al. (1996). Forested wetlands are generally further up in the watershed and are less significant in explaining the variability in sediment contaminant levels. The ability to delineate between forested wetlands and forests is a possible limitation with the land cover data set. However, forests only entered one model (organics, combination F), and this was not a best model.

Sediment metals concentrations increase also with increasing inputs from point source discharges in the

watershed. Data were not available for point source input of organic contaminants. It is expected that point source inputs of organics would be important. Not including these inputs in the regressions may partially explain the lower overall *R*<sup>2</sup> compared with the metals model. Other reasons include the high variability in the organics values and the large number of concentrations below detectable levels.

Because of the different sediment characteristics and dynamics of estuarine systems across the mid-Atlantic and southern New England regions, watershed-estuarine processes were shown to be important in describing the quantitative linkages. These processes mitigate the effect of the watershed stressors on sediment contaminant concentrations. In every case the best models included at least one of the following: sediment grain size, TOC concentration, and estuarine hydrology. Results indicated that it was more important (based on *R*<sup>2</sup>) to include estuarine hydrology as a grouping variable (two-equation model, i.e., different coefficients for the independent variables based on the grouping) than as an additional additive independent variable (single-equation model). This implies that the estuarine dynamics quantitatively changes the linkage between the watershed stressors and sediment contaminant levels rather than just provide an additive factor over and above that provided by the stressors.

The importance in accounting for sediment characteristics and estuarine dynamics for reasonable predictions of sediment contaminant levels provides for a possible approach to answering the question posed in the title of the paper by Rose (2000), "Why are quantitative relationships between environmental quality and fish populations so elusive?" First of all, the intervening processes (such as surface water dynamics) need to be taken into account. Secondly, the distinction needs to be explicitly made between local factors (such as water and sediment characteristics) and remote factors (for example, land use and point sources) in exploring the development of quantitative relationships.

The overall best model (largest *R*<sup>2</sup>) was for sediment metals. This model included grain size and grouping by estuarine hydrology. In contrast, the best models for organics and total PAHs included TOC instead of grain size. This suggests that if linkages were to be explored with benthic community conditions, then it may be necessary to normalize the organic contaminants by the organic C content of the sediment. This would account

**Table 6. Coefficients of determinations (*R*<sup>2</sup>) for models with natural log transform of dependent variable (Eq. [2]) developed from multiple linear regression with stepwise selection on first principal component for metals (PC1-MET) and organics (PC1-ORG) and total PAHs as dependent variable. Results shown only for those models with normally distributed residuals.**

Dependent variable	Independent variable combinations						
	A	B	C	D	E	F	G
PC1-MET	0.74	0.67	0.51	NN†	0.79‡	0.67	0.7
PC1-ORG	0.48§	0.33	0.41	0.35	0.42	0.46§	0.5‡§
Total PAHs	0.42§	NN	0.37§	NN	NN	0.39§	0.46‡§

† Residuals were considered nonnormal at *p* ≤ 0.01 with Shapiro-Wilk test, designated as NN. All of linear functional form models (Eq. [1]) had nonnormal residuals. Independent variables were at *p* ≤ 0.15 significance level to enter in model.

‡ Model with largest *R*<sup>2</sup>.

§ Models not significantly different determined by ANOVA at *p* ≤ 0.05.

**Table 7. Coefficients for statistical models with natural log transform of dependent variable (Eq. [2]) developed using multiple linear regression with stepwise selection with first principal component for metals (PC1-MET) and organics (PC1-ORG) and total PAHs as dependent variables (models not significantly different, ANOVA at  $p \leq 0.05$ ).**

Dependent variable	Independent variable combination					
	E					
	Class 1 and 2	Class 3 and 4	F		G	
<b>Sediment metals</b>						
HYDROLOGY						
GLACIAL						
SICL	0.013	0.020				
TOC						
AREA						
PURB	1.49	0.98				
PAG						
PFOR						
PFWET		5.42				
PNFWET		-6.20				
FLOW	$1.2 \times 10^{-7}$					
PC1-INPUT	0.094					
<b>Sediment organics</b>						
HYDROLOGY						
GLACIAL						
SICL	-1.26					
TOC	0.50	0.54	0.345	0.60	0.37	
AREA						
PURB	2.83	6.42	12.68†	3.30	2.03	
PAG			9.97†			
PFOR		3.84	8.76†			
PFWET		36.82	8.20			
PFNWT	-8.18			-7.85		
<b>Sediment total PAHs</b>						
HYDROLOGY						
GLACIAL						
SICL	-1.12					
TOC	0.69	0.62	0.60	0.74	0.87	0.45
AREA						
PURB	3.05	2.49	1.95	3.86	3.93	
PAG						
PFOR						
PFWET						
PNFWET	-9.13	-9.43	-20.84	-6.78	-7.21	-18.53
FLOW						

† Variance inflation factor (VIF) &gt; 10.

for the partitioning to the organic C and the *effective* exposure to the organisms.

The land cover data set used for the analysis was based on 1973 aerial photos. Two limitations to use of this data set are (i) areas of rapid changes since 1973 and (ii) quality of wetlands delineation. Some coastal areas, particularly in New Jersey and Delaware, have experienced major land use changes between the acquisition of the aerial photos and the collection of the sediment data. There is no statistical procedure to deal with this, and it remains as a possible limitation to the study. Since the analysis in this study has been completed, a more recent land cover data set for the entire region has become available (Vogelmann et al., 1998). The statistical models developed in this study should be evaluated against the more recent land cover data set, and is the subject of ongoing work. The quality of the wetlands delineation was uneven across states because of the different photo interpreters. Percent area of non-forested wetlands was an important component of the models. However, the delineation of the nonforested wetlands in the aerial photography could be expected

to be less variable than for forested wetlands. This limitation can also be addressed by the ongoing work with the new land cover data set.

Classical parametric methods (i.e., multiple linear regression with stepwise selection) were used in this study to develop the statistical models. A limitation in the application of the classical methods for developing the statistical models is the possible transformation of variables that can be used. One approach for expanding beyond the functional forms used with the classical parametric methods, and which are the subject of further research, are deterministic-statistical models. These models involve specification of a general mathematical form relating independent to dependent variables (typically nonlinear), derived from separate studies or available in the literature. The parameters in the model are solved by methods such as nonlinear least squares. An example of this approach is the work by Smith et al. (1997) and Preston and Brakehill (1999) in development of a set of spatially referenced regression models for the evaluation of nutrient loading in watersheds. The functional form they used was a sum of exponentials, with nonlin-

ear regression analysis used to solve for the parameters. Results from the classical methods do provide direction for those processes that should be explored with these more detailed models.

The log transformation of the dependent variables provides statistical models that can be used for applications that involve predictions with new sets of independent variables. As an example, the models could be applied to the independent data set collected in small estuarine systems in 1997–1998 by the mid-Atlantic Integrated Assessment estuarine program (Strobel et al., 2000). These data would be used in a validation mode to test the statistical models developed in this study against an independent data set. Another example would be to use the models in a forecasting mode for evaluating possible management actions, under the assumption that the relationships would be valid in the future. Scenarios for future distributions of land cover for different land management options could be used to predict what sediment contamination might result from the management actions.

### SUMMARY

The question posed at the beginning of this paper was how does one determine “which, if any, of these landscape metrics are appropriate for predicting the impact of nonpoint pollution on estuarine receiving waters?” We chose to address this question by developing statistical models that relate the landscape metrics to estuarine sediment contamination levels. This approach was similar to that introduced in Comeleo et al. (1996). We addressed the identified limitations in that study by (i) using a consistent land cover classification data set for the entire geographic region that included a wetlands class; (ii) using triple the number of independent estuarine systems for the analysis ( $n = 75$ ); and (iii) extending the results from the relatively, low-relief coastal plain watersheds to those across a broad, diverse geographic region.

Results suggest that variation in sediment contamination levels across small estuarine systems in the mid-Atlantic and southern New England regions was related to the surrounding land cover composition and point source discharges of pollutants. The influence of these variables was mitigated by sediment characteristics and estuarine dynamics. The landscape metrics important for explaining the variation in sediment metals levels ( $R^2 = 0.72$ ) were the percent area of nonforested wetlands (negative contribution), percent area of urban land, and point source effluent volume and metals input (positive contributions). The metrics important for sediment organics levels ( $R^2 = 0.5$ ) and total PAHs ( $R^2 = 0.46$ ) were percent area of urban land (positive contribution) and percent area of nonforested wetlands (negative contribution). Incorporation of estuarine hydrology and sediment characteristics into the statistical models was necessary to generate reasonable  $R^2$ . The result that percent area of urban land is the major correlate with sediment contamination is consistent with the conclusion of a recent National Water Quality Inventory

(USEPA, 1995) that runoff from urban area is the prime source of water quality impairments in estuaries. The overall results from this study are indicative of how statistical models can be developed relating landscape metrics to estuarine sediment contamination.

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