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Contaminants in Stream Sediments From Seven United States Metropolitan Areas: Part I: Distribution in Relation to Urbanization

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Abstract Organic contaminants and trace elements were measured in bed sediments collected from streams in seven metropolitan study areas across the United States to assess concentrations in relation to urbanization. Polycyclic aromatic hydrocarbons, polychlorinated biphenyls, organochlorine pesticides, the pyrethroid insecticide bifenthrin, and several trace elements were significantly related to urbanization across study areas. Most contaminants (except bifenthrin, chromium, nickel) were significantly related to the total organic carbon (TOC) content of the sediments. Regression models explained 45–80 % of the variability in individual contaminant concentrations using degree of urbanization, sediment-TOC, and study-area indicator variables (which represent the combined influence of unknown factors, such as chemical use or release, that are not captured by available explanatory variables). The significance of one or more study-area indicator variables in all models indicates marked differences in contaminant levels among some study areas, even after accounting for the nationally modeled effects of urbanization and sediment-TOC. Mean probable effect concentration quotients (PECQs) were significantly related to urbanization. Trace elements were the major contributors to mean PECQs at undeveloped sites, whereas organic contaminants, especially bifenthrin, were the major contributors at highly urban sites. Pyrethroids, where detected, accounted for the largest share of the mean PECQ. Part 2 of this series (Kemble et al. 2012) evaluates sediment toxicity to amphipods and midge in relation to sediment chemistry.

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Watershed urbanization increases loadings of contaminants to aquatic ecosystems, and aquatic sediments are an important repository of hydrophobic contaminants. Organochlorine pesticides, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and trace elements are frequently monitored and found in urban stream and lake sediments (e.g., Lopes and Furlong 2001; Paul and Meyer 2001; Yunker et al. 2002; Beasley and Kneale 2004; Van Metre and Mahler 2005; Walsh et al. 2005; Wilson et al. 2005; Gilliom et al. 2006; Mahler et al. 2006; Echols et al. 2008; Foster and Cui 2008; Horowitz and Stephens 2008; Li et al. 2011). Pyrethroid insecticides recently have emerged as contaminants of concern in urban streams, with toxic concentrations reported in sediments of small urban streams (e.g., Holmes et al. 2008; Hintzen et al. 2009; Weston et al. 2011), especially near stormwater or sewage discharges (e.g., Weston et al. 2005; Amweg et al. 2006; Ding et al. 2010).

Most studies of contaminants in urban streams have focused on a limited range of urban contaminants in highly urbanized areas either locally (e.g., reviews by Walsh et al. 2005; Paul and Meyer 2001) or nationally (Wong et al. 2000; Lopes and Furlong 2001; Gilliom et al. 2006; Horowitz and Stephens 2008). Little is known, however, about how contaminant mixtures change in occurrence and composition as urban development progresses from low to high density. A few studies have examined selected contaminants in stream sediment at sites ranging from remote or rural to urban within a localized area or region. Examples include PAHs and PCBs along the Potomac River (Foster and Cui 2008); PAHs in the Fraser River basin, Canada (Yunker et al. 2002); PAHs in estuarine basins in South Carolina (Van Dolah et al. 2008); and PAHs and trace elements in Yorkshire, UK (Beasley and Kneale 2004). Collectively, these studies show that sediment-dwelling benthic organisms in urban streams commonly are exposed to complex mixtures of contaminants in sediment.

Recent studies conducted by the United States Geological Survey (USGS) of water chemistry and ecological community characteristics at stream sites, along a gradient from undeveloped to highly urbanized watersheds, showed that the responses to urbanization varied among metropolitan areas (e.g., Sprague and Nowell 2008; Brown et al. 2009; Coles et al. 2009, 2012). The present study investigates the effects of urbanization on stream sediment contamination and toxicity. This article is the first of a two-part series evaluating the occurrence, composition, and toxicity of contaminant mixtures in bed sediment sampled at 98 total streams along urban gradients within seven metropolitan areas across the United States in 2007. The objectives of the overall study were to (1) determine contaminants in stream sediment under nonpoint source conditions in streams ranging from undeveloped to highly urbanized watersheds (basins) within each metropolitan area; (2) measure the toxicity of these sediments to amphipods and midge; (3) assess occurrence and toxicity in relation to urbanization; (4) evaluate relationships between sediment toxicity and sediment chemistry; and (5) compare results among the seven metropolitan areas. This article (part 1 of the series) characterizes five classes of sediment contaminants—including pyrethroid insecticides—in relation to urbanization and other factors; evaluates sediment contamination in the context of sediment-quality guidelines (SQG); and compares these results among the seven metropolitan areas studied. Part 2 of this series (Kemble et al. 2012) characterizes the toxicity of the sampled sediments to amphipods and midge in whole-sediment toxicity tests and evaluates relationships between sediment contamination and sediment toxicity.

Methods

Study Area and Site Selection

Seven metropolitan areas (study areas) were sampled: Atlanta (GA); Boston (MA, NH); Milwaukee–Green Bay (WI [Milwaukee]); Dallas–Fort Worth (TX [Dallas]); Denver (CO, WY); Salt Lake City (UT); and Seattle–Tacoma (WA [Seattle]; Fig. 1). These study areas vary with respect to ecoregion, climate, geology and soil properties, streamflow characteristics, and pre-urban land cover (Table 1). A total of 98 stream sites were sampled during 2007, with 12–14 sites in most study areas (see [Table 1] in Kemble et al. 2012). With few exceptions, sampling locations were wadeable streams in small- to moderate-sized basins (median 50 km^2; see [Figs. 2–8] in Moran et al. 2012). A site is defined as a reach of a wadeable stream that extends 20 times the average wetted channel width, or a minimum of 150 m, and that is void of significant inflows or outflows over its length (Moran et al. 2012).

Within each study area, sampling sites were selected to span a gradient of basin urbanization, from minimally to highly urbanized, while minimizing variability in natural environmental setting (ecoregion, climate, topography, soil type, stream-flow characteristics, etc.). In this way, natural factors would minimally confound the interpretation of response along the urban gradient (McMahon and Cuffney 2000). As a result, characteristics of the natural environmental settings varied substantially among study areas (Table 1) but comparatively less within a study area. For example, Atlanta and Seattle both have forested background land cover and high precipitation; however, Seattle basins have sandier, more permeable soil, higher latitude, and predominantly quaternary glacial deposits, whereas Atlanta basins have comparatively higher air temperatures and contain predominantly sedimentary rock. Additional details are in Moran et al. (2012).

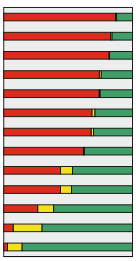
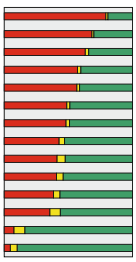
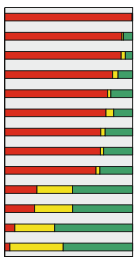
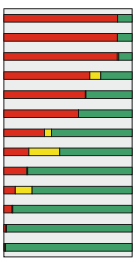
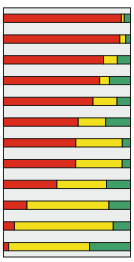
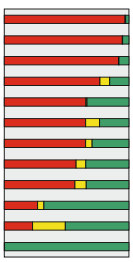
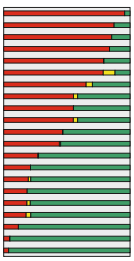
Ancillary Data for Study Sites and Basins

The drainage basin of each site was characterized using nationwide geographic information system (GIS) data sources as described in Moran et al. (2012). Briefly, each basin characteristic was determined by clipping the national coverage for that data type to the basin boundaries for each site. Streams were based on the USGS National Hydrography Dataset (NHD) 1:100,000-scale stream set (USGS and United States Environmental Protection Agency [USEPA] 2003). GIS data types and sources are listed in Tables S1.1 and S1.3.

Several GIS-derived measures of urbanization at the basin scale were used as generic surrogates for

Fig. 1 Map of the seven metropolitan study areas

Table 1 Selected environmental characteristics (range in values) for sites within the seven metropolitan study areas

Study area	ATL	BOS	DAL	DEN	MGB	SLC	SEA
Land use at individual sites along the urban gradient (Urban, Agriculture, Undeveloped)							
Ecoregion[a]	ET Forest	ET Forest	G Plains	G Plains	ET Forest	NA Desert	MWC Forest
Dominant geology	Sed	Gr, Sed, Gn	Sed	Sed, Gn, Gr	Sed	Qu, Sed	Qu, Vol
Mean annual air temp (°C)	15.6–16.5	7.8–9.9	17.5–18.5	0.2–9.7	6.8–8.7	6.7–11.2	9.8–11.0
Mean annual precip (cm)	132–140	112–128	90–105	40–95	77–89	41–84	98–166
Site elevation (meters)	216–281	1–122	93–167	1,395–2,444	186–232	1,291–1,664	5–104
Sand in soil (%)	34–36	48–65	9–35	25–27	10–26	27–56	36–61

ATL Atlanta, *BOS* Boston, *cm* centimeter, *DAL* Dallas, *DEN* Denver, *MGB* Milwaukee-Green Bay, *precip* precipitation, *SEA* Seattle, *SLC* Salt Lake City, *temp* temperature, *°C* degrees Celsius, *%* percent, *Gn* gneiss, *Gr* granitic, *Qu* quaternary, *Sed* sedimentary, *Vol* volcanic

[a] Level I ecoregion: *ET forest* Eastern temperate forest, *G plains* Great plains, *MWC forest* Marine west coast forest, *NA desert* North American desert, One Denver site is in North west forested mountains

urban-contaminant sources: urban land use/land cover (from 2001 National Land Cover Database [NLCD]; USGS 2008), population density (from the 2000 Census; SILVIS Laboratory 2008), road density (from 2000 Census Topologically Integrated Geographic Encoding and Reference [TIGER] line roads; GeoLytics 2001), and impervious surface (from 1-km National Oceanic and Atmospheric Administration data; see Moran et al. 2012). A surrogate for urban insecticide use called the termite-urban score was used for pesticides. This score was derived from urban land cover in the basin weighted by the subterranean termite-density zone (Beal et al. 1994) as described in Nowell et al. 2009; the types of urban land-cover data used for currently used pesticides (pyrethroids) and historically used

pesticides (organochlorines) were 2001 NLCD (USGS 2008) and 1992 enhanced NLCD data (Nakagaki et al. 2007), respectively. This termite-urban score is expected to reflect use to control subterranean termites and other urban insects that have similar geographic distributions to that of termites (i.e., greater density in the southeast and parts of California; Fig. S3.1), such as fire ants (Korzukhin et al. 2001; also see Supplementary Information S3). Urban influence local to the site was represented by 2001 NLCD urban land cover within the main-stem buffer (within 800 m of the stream) or riparian buffer (within 100 m of the stream) and by the number of road-stream intersections in the stream segment (within 100 m of the stream for a length of approximately 2000 m upstream from the sampling site [from Census 2000 TIGER roads and NHD streams]). For two sites in the Denver study area (Cherry and Bear Creeks), basin-scale land-use data underestimated the urban contribution to water flow at the sampling sites because water from undeveloped upper portions of the basin was often diverted or impounded (Sprague et al. 2006; also see Table S1.2). These sites were included in model fitting and data analyses but are noted as outliers in some analyses. Some analyses were performed on a subset of sites, comparing sites classified as highly urban (>50 % urban land cover [using 2001 NLCD data]) to those classified as undeveloped (<5 % urban).

Natural environmental setting was represented by multiple basin characteristics related to ecoregion, landscape and topography, climate, hydrology, geology, and soil properties (Table 1; Tables S1.1 and S1.3). In addition, study-area indicator variables were created for use as dummy variables in regression models. An indicator variable was created for each of the seven study areas; each variable was populated for a given site by "1" or "0" depending on whether the site is located in that study area. For example, each site in Atlanta would have a value of "1" for the Atlanta study area variable and a value of "0" for the other six study area variables. Study-area indicator variables were used in regression analysis to evaluate the presence of unknown natural or anthropogenic factors that are unique to individual study areas, but that are not represented by available explanatory variables.

Sample Collection and Processing

One bed sediment sample was collected from each of the 98 sites between May and September 2007 corresponding to seasonal low-flow conditions within each study area. Each sample was a composite of grab samples of the top 2 cm of sediment from multiple depositional zones within the stream reach. The composite sample was homogenized on site, and subsamples were sieved in the field to <2 mm (to remove gravel) for organic contaminant analyses and toxicity testing and to <63 μm (to remove sand and gravel) for trace element analyses (Moran et al. 2012).

Chemical Analysis

Sediment samples were analyzed for trace elements, PAHs, organochlorine pesticides, PCBs, and pyrethroid insecticides. Pyrethroids were analyzed by the USGS Pesticide Fate Research Project in Sacramento, CA, and all other contaminants were analyzed by the USGS National Water Quality Laboratory in Denver, CO (Moran et al. 2012). Analytical methods and quality control are described in Supporting Information S2. Briefly, sediment extracts were subjected to various clean-up and isolation procedures, then analyzed for organic contaminants by gas chromatography with electron-capture (organochlorine compounds) and mass spectrometric (PAHs and pyrethroids) detection. For trace elements, sediments were subjected to strong acid digestion and analyzed by hydride generation atomic absorption spectrometry (AAS) for arsenic, cold vapor AAS for mercury, and inductively-coupled plasma-atomic emission spectrometry for all other elements. Trace element analyses generated total (≥95 % of the element present) rather than total-recoverable concentrations (Horowitz and Stephens 2008). TOC in sediment was measured in both the 2 mm- and 63 μm-size fractions.

Toxicity Tests

Whole-sediment toxicity tests were conducted with the amphipod *Hyalella azteca* (28-day exposures) and the midge *Chironomus dilutus* (10-day exposures) according to established methods (USEPA 2000; American Society for Testing and Materials 2011). End points were survival, average weight, and total biomass of test organisms determined at the end of the exposures. Details are provided in part 2 of this series (Kemble et al. 2012).

Data Analysis

Statistical analyses were performed using TIBCO Spotfire S+ (TIBCO Software 2008 [http://spotfire.tibco.com/en.aspx]) or Data Desk 6.1 (Data Description 2011 [http://www.datadesk.com/products/data_analysis/datadesk/]). Relationships of contaminants to urbanization measures were assessed using Spearman's rank correlation analysis and linear least-squares regression with ranked concentrations. Differences in contaminant concentrations among study areas or other categorical variables were tested using nonparametric tests of association, including Kruskal–Wallis and Tukey multiple comparison tests. Censored values (i.e., nondetections expressed as lower than the reporting level) were ranked as ties below the lowest detected concentration. Of pyrethroids in the present

study, only bifenthrin was detected frequently enough to be included in statistical analyses.

Multiple regression models were used to evaluate the relationship of contaminant concentrations to urbanization, sediment characteristics, and other factors. Two approaches were used to evaluate the co-occurrence of contaminants in sediment: (1) principal components analysis (PCA), which evaluates associations among contaminants in a sample mixture; and (2) the mean probable effect concentration quotient (PECQ) procedure, which weights the concentration of each contaminant in a sample mixture by its relative toxicity to benthic organisms to obtain an overall indicator of predicted toxicity for the sample mixture. Concentrations of individual contaminants and classes also were compared with SQGs—specifically, probable effect concentrations (PECs) and equilibrium-partitioning sediment benchmarks (ESBs). Kemble et al. (2012) describe how various SQGs were calculated.

Regression Models

Multiple linear regression models were developed for 14 contaminants: total PAHs, total DDT, total chlordane, total PCBs (each defined in Table 2), dieldrin, bifenthrin, and aluminum-normalized concentrations of arsenic, cadmium, chromium, copper, lead, mercury, nickel, and zinc. Maximum likelihood (Tobit) methods appropriate for data with censoring were used and implemented in the survreg

Table 2 Explanatory variables that significantly explained contaminant concentrations in Tobit regression models and the percentage of variability explained by the models

Contaminant[a] detection frequency (%)	Explanatory variables in regression model[b]								pR^2: variability explained by		
	Urban surrogate variable (s)	TOC	Study-area indicators[c]								
			ATL	BOS	DAL	DEN	MGB	SLC	SEA	Urban only[d]	Urban + TOC[e]	Final model[f]
Bifenthrin (41)	TU_B							+	–	0.27	0.27	0.42
Dieldrin (41)	TU_B, U_L	TOC						+		0.70	0.72	0.75
Tot-chlordane (47)	U_B, U_L	TOC			–		–		–	0.32	0.58	0.71
Tot-DDT (44)	U_B	TOC	–		–				–	0.17	0.54	0.65
Tot-PAH (100)	U_B	TOC	+	+		+	+			0.23	0.39	0.67
Tot-PCB (47)	U_B	TOC			–					0.21	0.53	0.62
As/Al (100)	U_B	TOC	–	+		–				0.06	0.26	0.55
Cd/Al (100)	U_B, U_L	TOC	–					+	–	0.24	0.50	0.79
Cr/Al (100)		**TOC**		**+**	**+**		**+**	**+**	**+**		**0.11**	**0.45**
Cu/Al (100)	U_B	TOC	–		–			+	–	0.14	0.37	0.61
Pb/Al (100)	U_B, U_L	TOC		+		+		+		0.27	0.57	0.75
Hg/Al (97)	U_B, U_L	TOC		+				+	+	0.11	0.48	0.59
Ni/Al (100)	**U_B**			**+**	**+**		**+**	**+**	**+**	**0.06**	**0.06**	**0.71**
Zn/Al (100)	U_B, U_L	TOC						+	–	0.33	0.58	0.71

Regression coefficients for all urban variables and TOC were positive. The sign of the coefficient is shown for study-area variables: + = positive; – = negative. Bolded text indicates that models did not successfully explain contaminant variability. Blank cells indicate that this variable was not selected in the model

ATL Atlanta; *BOS* Boston; U_B basin-scale urban (medium-to-highly urban land in basin); TU_B basin-scale termite-urban score; *DAL* Dallas; *DEN* Denver; U_L local-scale urban (number of roads-stream intersections in stream segment); *MGB* Milwaukee-Green Bay; *SEA* Seattle; *SLC* Salt Lake City; *Tot* total

[a] Tot-chlordane = sum of cis-chlordane, trans-chlordane, and trans-nonachlor; Tot-DDT = sum of *p*,*p*′-DDT, *p*,*p*′-DDD, and *p*,*p*′-DDE; Tot-PAH = sum of acenaphthene, acenaphthylene, anthracene, fluorene, naphthalene, phenanthrene, benz(a)anthracene, dibenz(a,h)anthracene, benzo(a)pyrene, chrysene, fluoranthene, and pyrene; Tot-PCB = sum of Aroclors 1254, 1260, 1016, 1242

[b] All selected variables have $p < 0.05$. Blank cells indicate that variable was not selected

[c] + (positive coefficient) = study area has higher concentrations than expected from urbanization and/or TOC; – (negative coefficient) = study area has lower than expected concentrations

[d] For preliminary model containing urban variables only, if any were selected by the stepwise AIC procedure

[e] For preliminary model containing urban and TOC variables only, if any were selected by the stepwise AIC procedure

[f] For final model containing urban, TOC, and/or study area variables

procedure (Therneau 1999) in the TIBCO Spotfire S+ program. Model development is described in Supplementary Information S3. Briefly, models were developed using a stepwise procedure similar to stepwise regression, except that the Akaike information criterion (AIC) (Venables and Ripley 1999) was used to select explanatory variables. Explanatory variables were added to the model in stages, and the AIC-based procedure was applied during each stage to select explanatory variables that produced the model that best described the response variable data.

Response variables and some explanatory variables were log-transformed to minimize deviations from Tobit-method assumptions. Potential explanatory variables included grain size and TOC measured in sediment samples (which affect contaminant sorption to sediments); basin- and local-scale measures of urbanization (generic surrogates for urban sources); a termite-urban score (a surrogate for urban use to control termites and other urban insects with similar geographic distributions); natural basin characteristics (which may affect contaminant transport to streams); and study-area indicator variables (Table S1.3). The latter are implemented as regression dummy variables and used to identify study areas that departed significantly from the national model relating contaminant concentrations to sediment and basin characteristics for each site.

Exploratory regression models were run to test the effect of normalizing the response variables (to aluminum or titanium for trace elements or to sediment-TOC for organic contaminants) and to identify the most effective explanatory variables in explaining contaminant concentrations. Trace elements were normalized to aluminum because this increased the variance explained by the model and strengthened the relationship to urbanization compared with dry-weight or titanium-normalized trace element concentrations. The relative proportion of metals and aluminum in crustal material is fairly constant, but aluminum has relatively small inputs from anthropogenic sources (Schropp and Windom 1988). Contaminant concentrations were not normalized by sediment-TOC; instead, sediment-TOC was considered as an explanatory variable in contaminant models so that its contribution to explaining variability in contaminant concentrations could be quantified and compared with that of urbanization in accordance with the study objectives. The effect of sediment-TOC on bioavailability was considered as part of the PECQ approach. Variables that were not significant in exploratory models were dropped from consideration during final model development (see Supplementary Information S3). These dropped explanatory variables included all natural basin characteristics (e.g., representing soil properties, hydrology, and climate) and the percentage of sand, silt, and clay measured in the sediment sample.

In final models, explanatory variables were added to the model and evaluated in three stages: (1) basin-scale urban variables and measured characteristics of sediment samples, (2) local-scale urban variables, and (3) study-area indicator variables (Table S1.3). Regression models took the following form:

$$\log X = C + (k_1 \times \text{Urban}_{\text{basin}}) + (k_2 \times \text{Urban}_{\text{local}}) + (k_3 \times \text{Sed}_{oc}) + (k_{\text{i}} \times \text{Study area}_i) + \cdots (k_6 \times \text{Study area}_6)$$

where X is the contaminant concentration; C is the intercept; $\text{Urban}_{\text{basin}}$ and $\text{Urban}_{\text{local}}$ are urbanization variables at the basin and local scale, respectively; Sed_{oc} is sediment-TOC; and Study area_i is the ith study-area indicator variable. When a study-area indicator variable was selected in a final model, it signified that some unknown combination of natural and/or anthropogenic factors characteristic of that study area caused the particular contaminant to be higher or lower than at other sites with similar values of sediment-TOC and urbanization. Model goodness of fit was assessed using the pseudo-R^2 (pR^2), which estimates the proportion of variation in the response variable explained by the Tobit model (Laitila 1993), and the scale, which is a low-biased estimate of the SD of the residual error (Aitkin 1981).

PCA

PCA was performed on the ranks of concentrations for the 44 most commonly detected, toxicologically important contaminants—31 organic analytes, eight trace elements, aluminum, titanium, and three carbon measurements. Organic analytes were included if they either (1) were detected in ≥30 % of samples, (2) had PEC values and were detected in ≥20 % of samples, or (3) were components of summed contaminant groups with PEC values (e.g., total PCBs). Only trace elements with PEC values were included. Titanium, aluminum, and carbon were included because these sediment characteristics are sometimes used to normalize contaminant concentrations.

Each contaminant has a loading for each component such that loadings reflect the correlation (shared variance) of the contaminant with that component. Loadings >0.7 were considered high ($0.7^2 = 0.49$, so approximately half the variance in the contaminant was associated with that component). Loadings >0.5 but <0.7 were considered moderate ($0.5^2 = 0.25$, so between a quarter and approximately half of the variance was associated with that component). An individual study area was considered to be high or low, relative to other study areas, in its association with a given component, if its component scores were

significantly higher or lower (respectively) than at least three of the other six study areas. More detail on PC analysis is provided in Supporting Information S4.

PECs

PECs are empirically derived SQGs developed on the basis of correlations between measured chemical concentrations and observed toxicity in field sediments. The PEC defines a contaminant concentration above which adverse effects on benthic organisms are expected to occur more often than not (MacDonald et al. 2000). The PECQ for an individual contaminant is its measured concentration divided by its PEC and thus represents concentration weighted by relative toxicity. The overall mean PECQ for a sample mixture is the average of PECQ values for all contaminants in the mixture.

An overall mean PECQ was determined for five classes of contaminants (mean PECQ-5) in a given sample—trace elements, PAHs, PCBs, organochlorine pesticides, and pyrethroid insecticides—after normalizing contaminant concentrations in the sample by sediment-TOC. Because it was important to include pyrethroids, although published PECs were not available for these compounds, similar guidelines to PECs were estimated for pyrethroids using sediment-toxicity data from the literature (Kemble et al. 2012; Moran et al. 2012).

The overall mean PECQ-5 was calculated by averaging the following: (1) the mean PECQ of arsenic, cadmium, chromium, copper, lead, nickel, and zinc; (2) the PECQ of total PAHs; (3) the PECQ of total PCBs; (4) the mean PECQ of DDE, dieldrin, and total chlordane; and (5) a toxicity quotient for pyrethroids calculated one of three ways. Depending on how pyrethroids were incorporated, the mean PECQ-5 is either: *mean PECQ-5M* (in which the pyrethroid term is the mean toxicity quotient of the four pyrethroids with available toxicity values, bifenthrin, cypermethrin, λ-cyhalothrin, and permethrin); *mean PECQ-5P* (in which the pyrethroid term is the summed toxicity quotients of bifenthrin, cypermethrin, λ-cyhalothrin, and permethrin); or *mean PECQ-5B* (in which pyrethroids are represented by the toxicity quotient for bifenthrin, which was the most commonly detected pyrethroid in this study). In all three cases, pyrethroid nondetections were assumed equal to zero because pyrethroid reporting levels were not far below their toxicity thresholds, whereas other contaminant nondetections were assumed to be half of the reporting level per the original PECQ methodology (Ingersoll et al. 2001).

Data analyses were performed three ways using all three versions of the mean PECQ-5, and the results were similar (as discussed later in the text and in Kemble et al. 2012). The mean PECQ-5B was selected in the current article to illustrate the potential toxicity of the five contaminant classes. The mean PECQ-5M probably reflects a *minimum* contribution of pyrethroids, relative to other contaminants, in terms of toxicity-weighted concentrations in sample mixtures because for the PECQ-5M approach: (1) toxicity quotients are averaged for pyrethroids, which have a common mechanism of action and are expected to have additive toxicity (Trimble et al. 2009); and (2) pyrethroid nondetections were assumed equal to zero instead of half the reporting level. In contrast, the mean PECQ-5P reflects the additive toxicity of pyrethroid mixtures because toxicity quotients for pyrethroids are summed. However, the mean PECQ-5P may underestimate the relative contribution of organochlorine pesticides (because if one or two component pesticides were not detected, then averaging PECQ values for the three component pesticides would actually lower the contribution of the detected pesticide to the potential toxicity of the mixture) compared with pyrethroids. In contrast, treating pyrethroid nondetections as zero has the opposite effect, possibly underestimating the relative contribution of pyrethroids compared with the other contaminant classes. Finally, the mean PECQ-5B relies on measured concentrations of bifenthrin alone, which was the most commonly detected pyrethroid, to represent the relative contribution of pyrethroids to the potential toxicity of the mixture. The mean PECQ-5B will underestimate the pyrethroid contribution to the potential toxicity in samples with detectable concentrations of cypermethrin, λ-cyhalothrin, or permethrin, but this occurred in only 15 % of samples, in which bifenthrin accounted for 12–96 % (median 65 %) of the summed pyrethroid toxicity quotients. The mean PECQ-5B was more effective in explaining measured toxicity to amphipods than either the mean PECQ-5M or mean PECQ-5P (Kemble et al. 2012).

ESBs

Measured concentrations of dieldrin and PAHs were compared with ESBs, which are mechanistically derived SQGs from the USEPA (2002, 2003). Based on equilibrium-partitioning theory, the ESBs assume that the bioavailability of nonionic organic compounds in sediment is controlled by sorption to sediment-TOC and incorporates select toxic-effect concentrations in porewater (i.e., final chronic values). Measured concentrations of contaminants are normalized by sediment-TOC before comparison with ESBs. Of the organics analyzed in this study, ESBs for only dieldrin and total PAHs are available. Because PAH compounds have a common mode of action, their toxicity values are integrated into an ESB toxic unit model ($\sum$ESBTU) that accounts for joint toxicity (see Kemble et al. 2012).

Potential Toxicity Thresholds

ESBs and PECQs can be used to predict toxicity associated with contaminants in field-collected samples. A dieldrin ESB quotient >1 or total PAH ∑ESBTU >1 indicates that benthic organisms in sediments may be unacceptably affected by dieldrin or PAHs, respectively. In addition, based on empirical associations of PECQ values with sediment toxicity occurring in field samples from past data sets, a high likelihood of toxicity is predicted for samples with PECQ values >1 for individual contaminants or an overall mean PECQ value >0.1 for sample mixtures (Ingersoll et al. 2001, 2005, 2009). Mean PECQs have been widely used in the literature to interpret toxicity data in field-collected sediments (e.g., Ingersoll et al. 2005) and to assess the biological significance of contaminants identified in studies that measured chemistry, but not toxicity, in field sediments (reviewed by Long et al. 2006).

Results and Discussion

Occurrence of Contaminants in Sediment

Although the 98 stream basins sampled ranged from mostly undeveloped to highly urbanized, organic contaminants were frequently detected and usually occurred as mixtures. One or more organic contaminants were detected in 98 % of sediment samples, ≥2 in 96 % of samples, and ≥10 in 78 % of samples; a median of 19 organic compounds were detected per sample. PAHs were detected in 98 % of samples and had the highest concentrations (Fig. 2). Total PCBs, dieldrin, total chlordane, total DDT, and bifenthrin were each detected in between 41 and 47 % of samples (Supporting Information Fig. S5.1). Organic carbon ranged from 0.2 to 17 % (median 3.4 % [mean 3.7 %]) in the 2-mm sieved sediment samples (Moran et al. 2012). Trace-element detections represent total concentrations in sediment; thus, detection frequencies for individual elements were high (95–100 %) for all elements except silver (11 %). Trace-element concentrations varied by element, study area, and site as shown in Fig. 3 for the eight trace elements with PECs.

On a dry-weight basis, contaminant mixtures in sediment samples were dominated by trace elements and PAHs, with much lower concentrations of PCBs and pesticides. For example, maximum concentrations were 465 mg/kg lead, 1,680 mg/kg zinc (Fig. 3), 80 mg/kg total PAHs, 882 µg/kg total PCBs, 102 µg/kg total DDT, and 11 µg/kg bifenthrin (Fig. 2). There were significant differences in contaminant concentrations among study areas (Figs. 2, 3). Concentrations were highest in samples from the Atlanta study area for dieldrin; in Dallas for bifenthrin; in Seattle for nickel; and in Boston for PAHs, PCBs, DDT, chlordane, arsenic, cadmium, and lead. Organic contaminant concentrations tended to be lower in Seattle than in most other study areas.

Although dry-weight concentrations were much lower for organic contaminants than trace elements, the reverse was true after concentrations were weighted by relative toxicity (i.e., divided by PECs) as shown by contributions of each contaminant class to the overall mean PECQ-5B (Fig. 4). As illustrated for organic contaminants in Atlanta, for example, normalization by sediment-TOC (Fig. 5b) can alter which sites have relatively higher total contaminant concentrations compared with dry-weight concentrations (Fig. 5a), but further weighting by relative toxicity (Fig. 5c) dramatically alters which contaminants are predicted to be more important in terms of potential toxicity to sediment-dwelling organisms. Despite their relatively low dry-weight concentrations (Fig. 2), pyrethroids substantially influenced the mean PECQ-5B at one or more sites in all study areas (Fig. 4). Dallas had the highest mean PECQ-5B values, largely due to bifenthrin (Fig. 4c). Pyrethroids also substantially influenced the mean PECQ-5M (at one or more sites in all study areas except Milwaukee [data not shown]) and the mean PECQ-5P (to a greater degree at some sites [data not shown] than shown in Fig. 4 for mean PECQ-5B).

Of the 44 contaminants subject to PCA, some contaminants tended to co-occur as evidenced by strong to moderate (>0.5) loadings on the same component (Table 3; Supporting Information S4). In PCA of ranked contaminant concentrations, five components were retained because (1) they had eigenvalues >1.0, indicating that each of these components accounted for a greater proportion of the variance than any one of the 44 individual contaminants in the original data set (Kaiser 1960); (2) they were hydrologically meaningful and had at least two high-loading contaminants; and (3) the scree test showed a linear progression for the higher components in explaining variance. All PAHs except one (2,6-dimethylnaphthalene) loaded strongly on the first component (PC1), which explained 39 % of the variability in the original contaminant concentrations (Table 3). Organochlorine pesticides, PCBs, and bifenthrin all loaded onto the second component (PC2), which explained 15 % of the variability in the original data. The next three components were composed of trace or major elements and explained 12 % (PC3 for lead, mercury, carbon, zinc, and copper), 7 % (PC4 for aluminum and titanium), and 6 % (PC5 for nickel and chromium) of the variability in the original data set. The single PAH not associated with PC1, 2,6-dimethylnaphthalene, is an alkylated two-ring PAH that is more characteristic of petrogenic than pyrogenic PAH sources (Neff et al. 2005); this was associated with PC3, as were carbon and organic

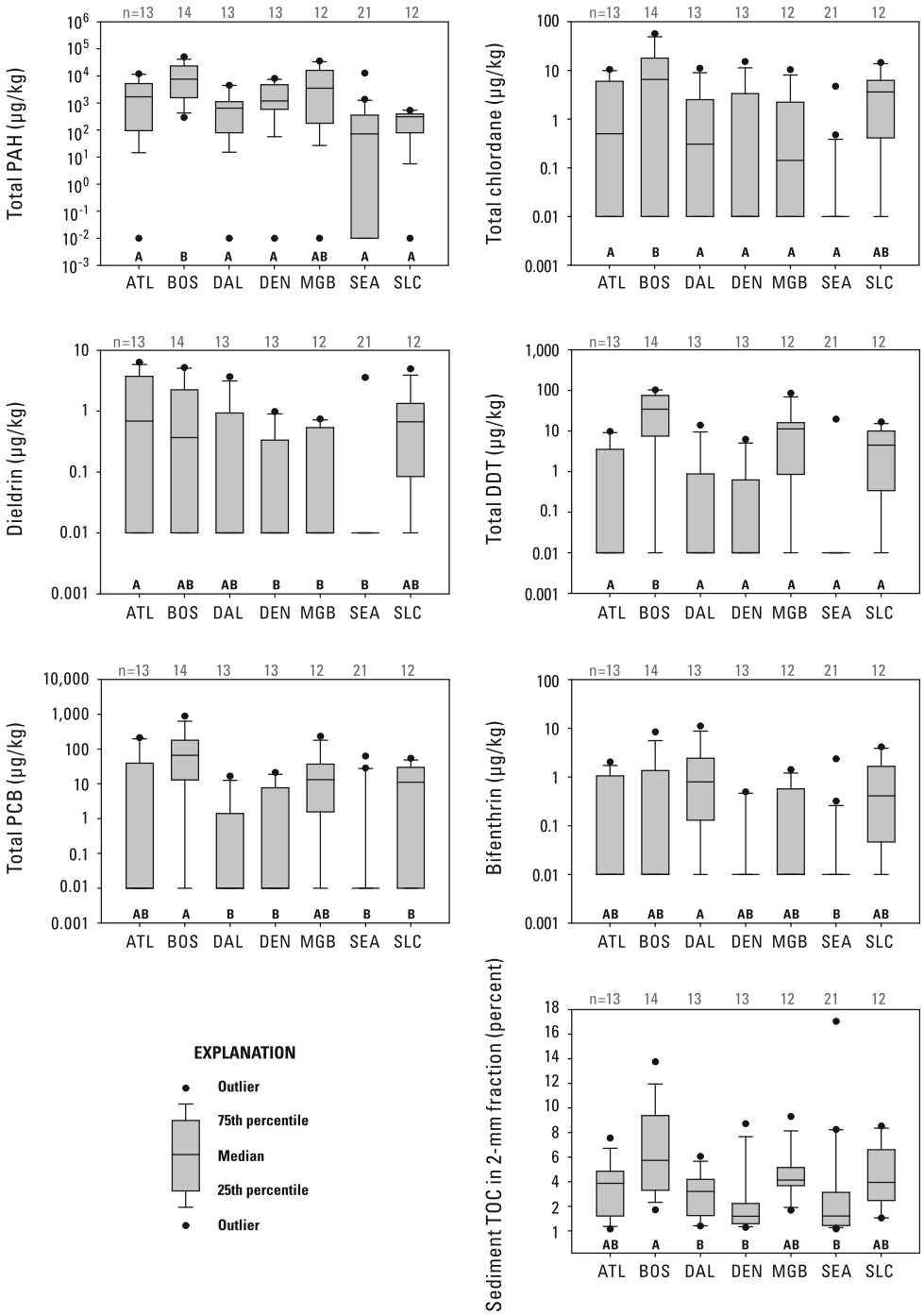


Fig. 2 Concentration distributions of organic contaminants and TOC in the 2-mm sediment fraction (dry weight) by study area. *Letters A,B* (*above x-axis*) indicate statistical significance; study areas with no letter in common are significantly different from one another (Tukey's test $p < 0.05$)

carbon from the <63-µm sediment fraction. Because PC4 and PC5 each explained relatively little variance in the original data set (≤7 %), they will not be discussed further.

Only those components associated with organic contaminants (PC1 and PC2) were significantly related to urbanization ($p < 0.01$; Table 3). PC1 scores (representing PAHs) were significantly higher for Boston, and significantly lower for Salt Lake City and Seattle, than at least three other study areas (Fig. S4.1). PC2 scores (pesticides and PCBs) tended to be highest for Salt Lake City and lowest for Seattle, whereas PC3 scores (selected trace elements) were highest for Boston and lowest for Dallas and Milwaukee (Fig. S4.1). Note that variability in PCA scores for a given study area included variability due to urbanization because sites within individual study areas range from <5–>80 % urban land use in the basin. Overall, the different components identified by the PCA reflect different sources for the associated contaminants as indicated by their chemical structures, likely origin, and relation of concentrations to land use. PAHs (PC1) have both

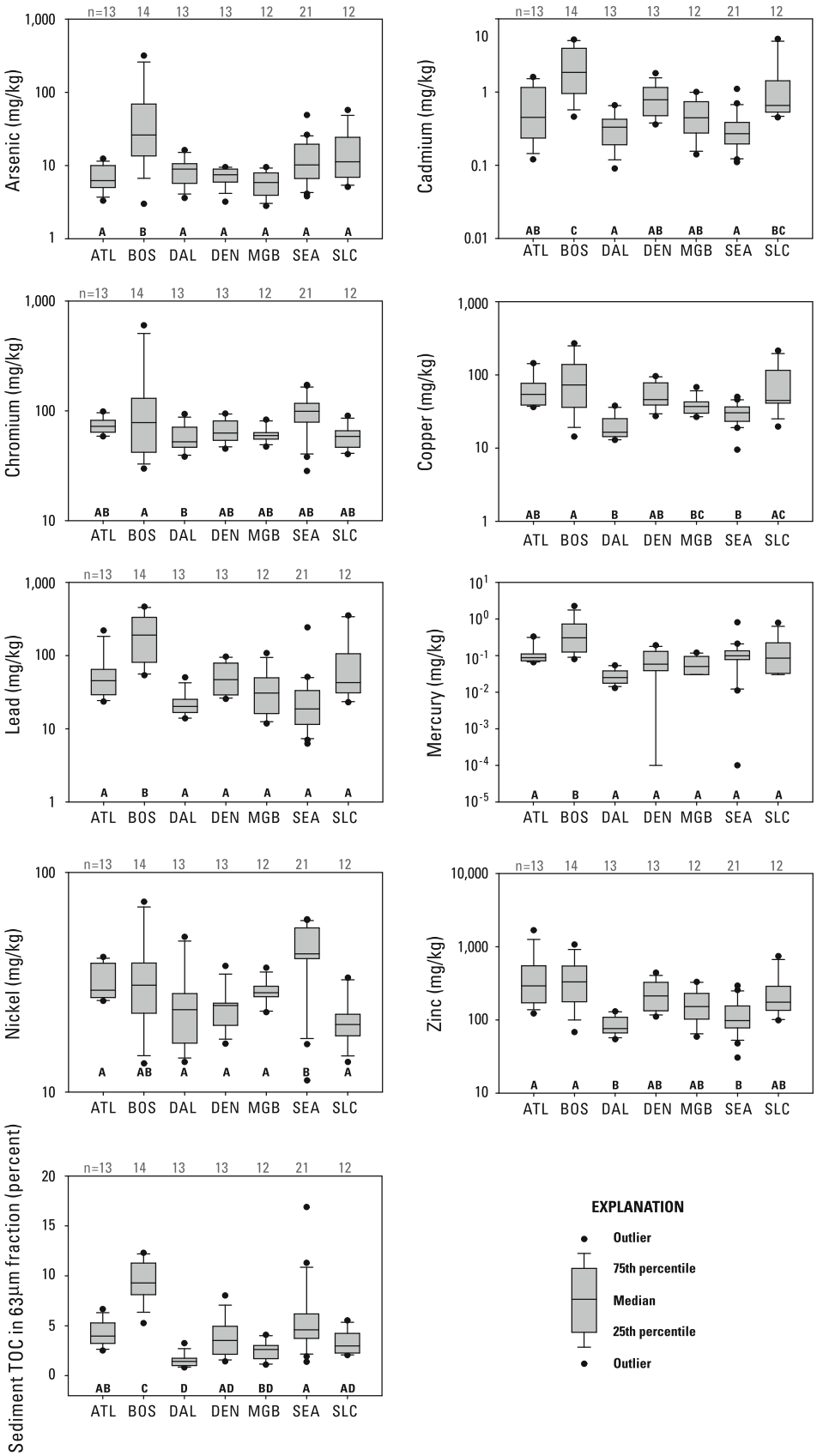


Fig. 3 Concentration distributions of selected trace elements and TOC in the 63-µm sediment fraction (dry weight) by study area. *Letters A–D* (*above x-axis*) indicate statistical significance; study areas with no letter in common are significantly different (Tukey's test $p < 0.05$)

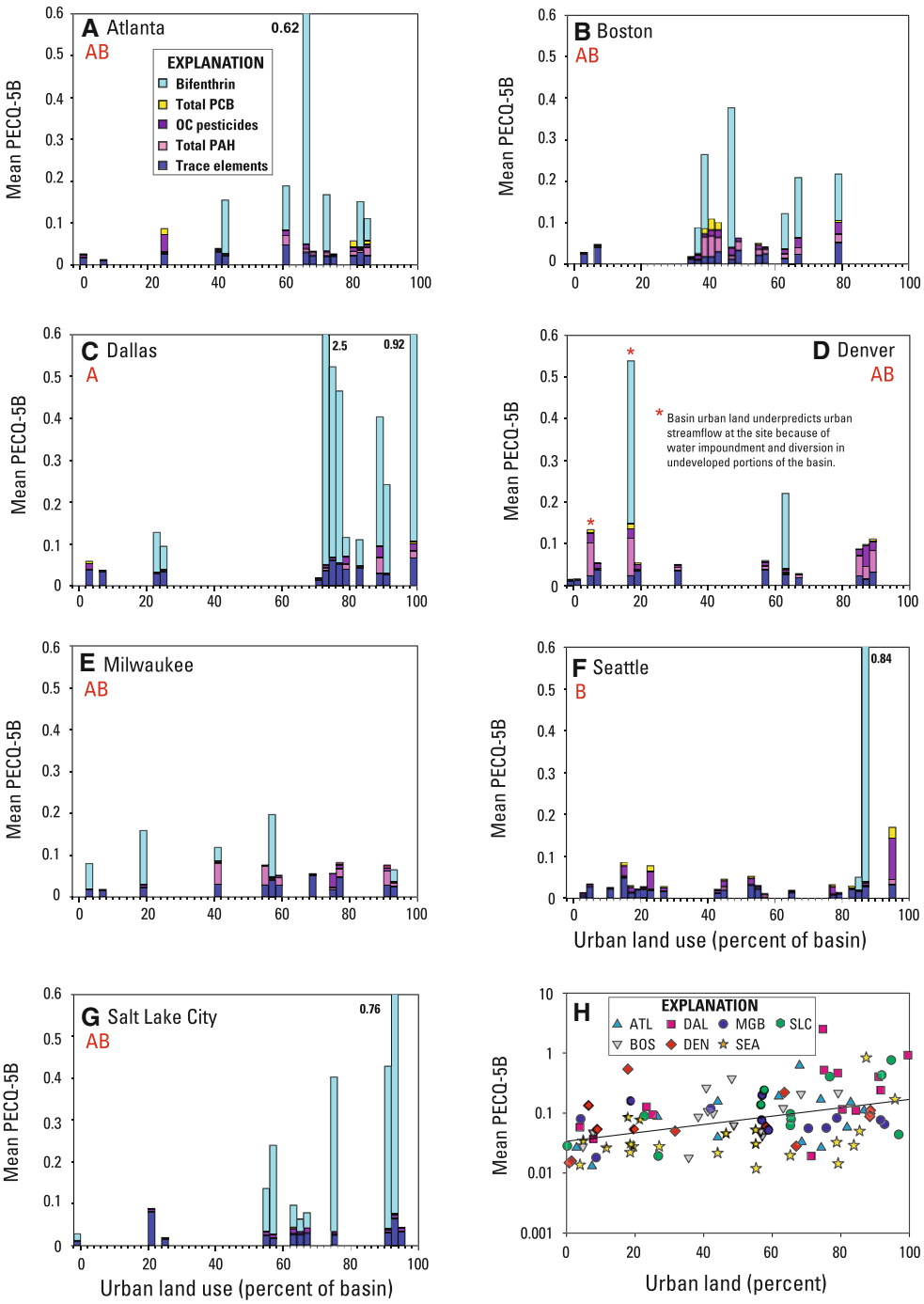


Fig. 4 Mean PECQ-5B in relation to urban land in the basin. **a**–**g** Contribution of five contaminant classes by study area. **h** Overall. *Red letters* (*upper left corner*) indicate whether study areas are significantly different from one another; study areas with no letters in common are significantly different (Tukey's test, $p < 0.05$)

natural sources (e.g., oil seeps) and anthropogenic sources (combustion products), whereas PCBs and pesticides (PC2) have exclusively synthetic sources. Trace elements (including those associated with PC3) tend to be ubiquitous in sediment, and their sources are typically natural (bedrock), but concentrations may increase from human activities, such as mining, incineration, or transportation. The top insecticides used in nonagricultural applications have changed over time. For example, the historically used organochlorine pesticides and chlorpyrifos were used until the early to mid-1980s (Nowell et al. 1999); diazinon, chlorpyrifos, malathion, and carbaryl were used during the 1990s (Aspelin 1997; Aspelin and Grube 1999; Donaldson et al 2002; Kiely et al. 2004); and pyrethroids, carbaryl, and malathion were used in 2007 (Grube et al. 2011). Notably, bifenthrin and the historically used organochlorine pesticides all had moderate to strong loadings on the same component (PC2), suggesting similar relative pest pressure among study areas, although the insecticides applied have changed over time.

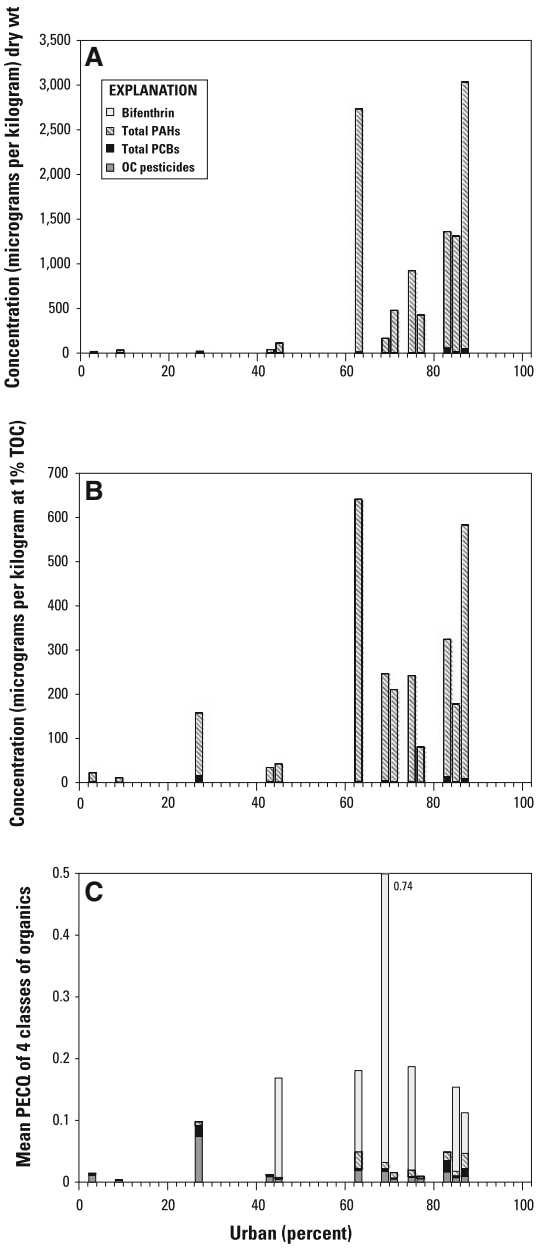


Fig. 5 Summed organic contaminant concentrations in Atlanta along the urban gradient. **a** Dry weight (μg/kg). **b** Normalized to sediment-TOC (μg/kg at 1 % sediment-TOC). **c** Normalized to sediment-TOC and by relative toxicity as mean PECQ of four classes of organics. Pyrethroids are represented by bifenthrin only

Nationally, concentrations of organochlorine pesticides and PCBs have decreased in hydrologic systems since their uses were discontinued in the 1970s (including most agricultural uses) and 1980s (termiticide use; Nowell et al. 1999) as evidenced by decreasing long-term trends in sediment cores from urbanized lake basins (Van Metre and Mahler 2005) and in fish from large rivers in the United States (Schmitt et al. 1990; Gilliom et al. 2006) and from the Great Lakes (Hickey et al. 2006). In sediment cores from urbanized lakes, decreasing trends outnumbered increasing trends for several trace elements, especially lead, chromium, cadmium, and nickel (Mahler et al. 2006). In contrast, PAHs have shown increasing trends in urban lake sediment cores (Van Metre and Mahler 2005). Although sales data for pyrethroids indicate increasing urban use of these insecticides (Spurlock and Lee 2008; Grube et al. 2011), insufficient data are available to evaluate trends in pyrethroid concentrations in urban stream sediments.

Relationships to Urbanization and Sediment Organic Carbon

All generic basin-scale urbanization measures (population density, urban land cover, road density, and impervious surface) were highly and significantly correlated with one another (Spearman rho ≥ 0.9, $p < 0.01$) with the termite-urban score (Spearman rho ≥ 0.8, $p < 0.01$) and with urban land cover at the local scale (within the main-stem and riparian buffers; Spearman rho ≥ 0.7, $p < 0.01$). For one local-scale urbanization measure, i.e., the number of road-stream intersections in the stream segment, correlations to the other urbanization measures were weaker (Spearman rho 0.2–0.3), but most were significant ($p > 0.05$) (see Table S6.1). Rank-transformed concentrations of all organic contaminants and four trace elements (cadmium, copper, lead, and zinc) were significantly ($p < 0.05$) and positively related to all basin-scale urbanization measures (Tables S6.2 and S6.3). For arsenic and mercury, urban relationships were weaker but significant for one or more urbanization measures. However, ranks of chromium and nickel concentrations were not significantly related to any urbanization measures (Table S6.3). Similarly, Horowitz and Stephens (2008) reported enriched concentrations of cadmium, copper, lead, mercury, and zinc in urbanized basins relative to baseline in sediment samples collected nationwide and weaker urban relationships for chromium, nickel, and arsenic. In the present study, aluminum normalization improved the relationships of all trace elements to urbanization, such that aluminum-normalized chromium and nickel were significantly related to one of seven urban measures (Table S6.3).

Concentrations of all organic contaminants, except bifenthrin, and all aluminum-normalized trace elements were significantly related to sediment-TOC (Tables S6.2 and S6.3). This is expected because both nonionic organic compounds and trace elements tend to partition to organic material in sediments (Di Toro et al. 1991; Mahony et al. 1996). The reason for the weak relationship of bifenthrin to sediment-TOC is unknown, but the influence of organic carbon could be obscured by regionally variable bifenthrin

Table 3 Contaminants loading onto the first three PCs from PCA of ranked concentrations of 44 contaminants to five components and significant relationships of PCs to urbanization and study areas

Component has:	PC 1: PAHs	PC 2: pesticides, PCBs	PC 3: TEs
High loadings (>0.7)	Benzo[k]fluoranthene	Dieldrin	Organic carbon (<63-μm fraction)
	Acenaphthene	*trans*-chlordane	Mercury
	Benzo[a]pyrene	*trans*-nonachlor	Lead
	Perylene	*cis*-chlordane	
	Benzo(b)fluoranthene		
	Benzo[ghi]perylene		
	Benz[a]anthracene		
	Chrysene		
	4H-cyclopenta-[def]phenanthrene		
	Phenanthrene		
	1-Methylphenanthrene		
	Fluoranthene		
	1-Methylpyrene		
	Benzo[e]pyrene		
	Acenaphthylene		
	Anthracene		
	Fluorene		
	Pyrene		
	Naphthalene		
Moderate loadings (0.5 to 0.7)		*p,p′*-DDE	Cadmium
		p,p′-DDT	Zinc
		Aroclor 1260	Copper
		p,p′-DDD	Carbon (<63-μm fraction)
		Aroclor 1254	2,6-Dimethyl-naphthalene
		Bifenthrin	Arsenic
		Aroclor 1042 + 1016	
Variance explained by PC (%)	39	15	12
Eigenvalue	17.1	6.6	5.3
Related to urban	Yes	Yes	No
High-loading study areas	BOS	SLC	BOS
Low-loading study areas	SLC, SEA	SEA	DAL, MGB

High and low study areas are significantly different from three or more of the other study areas (Tukey's test). Total variance explained by the first three PCs is 66 %. Two other components (not shown) contained aluminum and titanium (PC4) and chromium and nickel (PC5) and explained only 7 and 6 % of variability, respectively

BOS Boston; *DAL* Dallas-Fort Worth; *MGB* Milwaukee-Green Bay; *SEA* Seattle-Tacoma; *SLC* Salt Lake City; *TE* trace element

use patterns (Kuivila et al. 2012). Concentrations of TOC in the sediments were not significantly related to urbanization in the seven metropolitan areas sampled, although previous studies have reported enhanced sediment-TOC in urban streams (e.g., Horowitz and Stephens 2008). The reason is unknown, but regional differences in the concentrations (Fig. 2) and types of organic carbon present may play a role.

The overall mean PECQ-5B increased significantly with increasing urbanization (Fig. 4h). At highly urban sites (>50 % urban), pyrethroids accounted for an average of approximately 75 % of the mean PECQ-5B, all other organics combined approximately 11 %, and trace elements only approximately 13 % (Fig. 6a). At undeveloped sites (<5 % urban), mean PECQ-5B values were on average much lower, with trace elements accounting for an average of approximately 55 % and all organics approximately 45 % of the mean PECQ-5B (Fig. 6b).

Of six sediment-toxicity test end points measured in the present study (Kemble et al. 2012), two were significantly related to the percentage of urban land in the basin: survival and biomass of the amphipod *H. azteca* in 28-day tests ($p < 0.05$; Fig. 7). No relationship to urbanization was found for the weight of *H. azteca* in 28-day tests or for

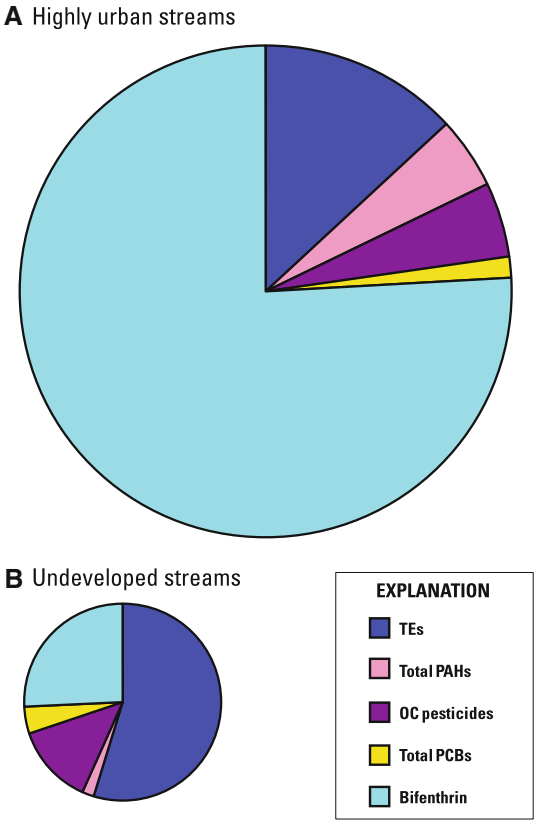


Fig. 6 Average contribution of five contaminant classes to the mean PECQ-5B in **a** 55 highly urban streams (>50 % urban land) and **b** nine undeveloped streams (<5 % urban land)

survival, biomass, or weight of the midge *C. dilutus* in 10-day tests. Relationships of toxicity end points to contaminant PECQs are described in Kemble et al. (2012).

Contaminant Responses to Urbanization and Other Factors

Multiple regression models were used to ascertain which explanatory variables best explained contaminant concentrations in the aggregated data set; the results are listed in Table 2. Of the 14 contaminant models, all but chromium contain a basin-scale urban variable; all but nickel and bifenthrin contain the sediment-TOC variable; and 6 models contain the local-scale urban variable (Table 2). Models for chromium and nickel were not successful because most of the variability explained was due to study-area variables (Table 2), so these models are not discussed further. For the 12 remaining contaminants, the urban variable(s) alone accounted for 6–70 % of variability in contaminant concentrations depending on the contaminant; addition of sediment-TOC increased the variability explained to 25–72 % (Table 2). Sediment-TOC alone accounted for 11–39 % of the variability (Table S3.2). The subsequent addition of study-area variables to the models increased the variability explained by a median of 11 % for organics and 27 % for trace elements. Study area variables with positive coefficients ("+" in Table 2) indicate that the modeled contaminant concentrations in these study areas were higher than expected on the basis of degree of urbanization and/or sediment-TOC; similarly, study-area variables with negative coefficients ("–" in Table 2) indicate lower concentrations than expected on the basis of urbanization and sediment-TOC. Regression coefficients for model equations are listed in Table S3.2.

Organic Contaminants

The models explained 62–76 % of the variability in concentrations for all organic contaminants except bifenthrin (42 %). For two of the four pesticides (dieldrin and bifenthrin but not DDT or chlordane), the termite-urban score was selected over a generic urban variable (Table 2). Although these pesticides were used to control other insects in addition to termites, the termite-urban score explained more of the variability in concentrations of dieldrin and bifenthrin than did urban land-use alone or any other generic urban surrogate. Historically, aldrin and its degradate, dieldrin, were used to control termites, moths, and insect-borne diseases, and as wood preservatives (USEPA 1992); chlordane was used for termite control and around homes and gardens (USEPA 1980); and DDT was

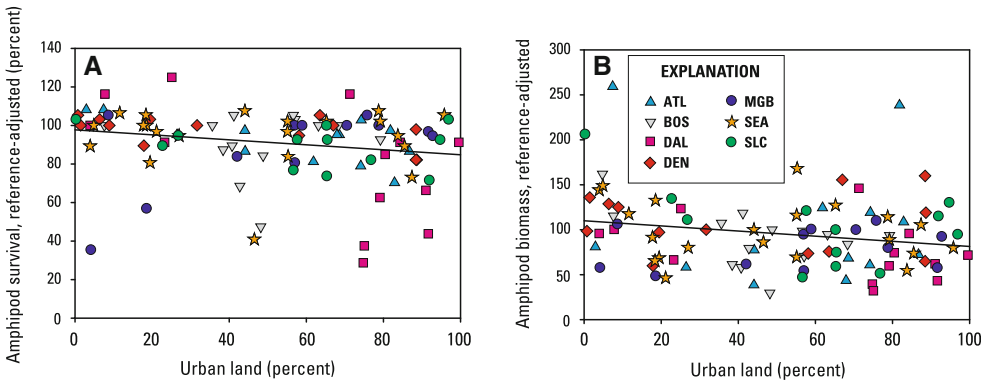


Fig. 7 The reference-adjusted survival (**a**) and biomass (**b**) of amphipods (*H. azteca*) in 28-day whole-sediment toxicity tests in relation to urban land in the basin.
a Survival = 97.6106–0.12898 (% urban), $r^2 = 0.05$.
b Biomass = 109.788–0.28596 (% urban), $r^2 = 0.05$

applied to control insect-borne diseases (USEPA 1992). Bifenthrin and other pyrethroids are commonly used in preconstruction soil treatments, structural applications, landscaping, mosquito control, and home and garden applications (Spurlock and Lee 2008; Moran and TenBrook 2011). For the dieldrin model, the termite-urban score by itself explained 70 % of the variability in dieldrin concentrations (Table 2). The single study area variable selected (Salt Lake City) had a positive coefficient, indicating that sites there had higher dieldrin concentrations than otherwise similar sites in other study areas, although the addition of this variable added only 3 % to the total variability explained by the dieldrin model, which was 75 % (Table 2). In contrast, the termite-urban score by itself explained only 27 % of the variability in bifenthrin concentrations, and the total variability explained by the model increased only to 42 % after study-area variables (Salt Lake City and Seattle) were added. Pyrethroid runoff into surface waters is more likely to occur from broadcast spraying onto impervious surfaces to control ants and other outdoor home and garden insects than from termiticide use, which typically occurs by underground injection or preconstruction treatment where there is little or no exposure of treated soil to water (Moran and TenBrook 2011). Although the termite-urban score is based on the geographic distribution of termites, this score also should represent pyrethroid use to control other urban insects with distributions similar to that of termites (e.g., fire ants; Korzukhin et al. 2001), but not insects with dissimilar distributions (e.g., carpenter ants; Klotz et al. 1999). The poorer performance of the bifenthrin model may reflect the fact that current pyrethroid use patterns are not fully represented by the termite-urban score (also see Supporting Information S3).

Regression models for total DDT, total chlordane, total PCBs, and total PAHs all contained the generic urban land-use variable and explained between 62 and 71 % of the variability in contaminant concentrations. Urbanized sites had significantly higher ratios of DDT/total-DDT and lower ratios of trans-nonachlor/total chlordane—which both are characteristic of recently deposited, less weathered sediments (Nowell et al. 1999)—and a higher proportion of combustion PAHs/all PAHs (Fig. S5.2).

Trace Elements

The four urban-influenced trace elements—cadmium, copper, lead, and zinc—had similar concentration distributions by study area, with concentrations being highest in Boston and lowest in Dallas and Seattle (Fig. 3). After addition of study-area variables to these four models, the total variability explained was 61–79 % (Table S3.2). Arsenic and mercury concentrations were highest in Boston, and the variability explained by their models was 55–59 % after the addition of study-area variables—including Boston—to correct for higher-than-expected concentrations.

Comparison of Study Areas

The inclusion of one or more study-area variables in all final models indicates significant differences among study areas that were not represented by available explanatory variables. Factors affecting contaminant concentrations within a particular region were not identified during regression modeling because study-area dummy variables (each of which represents the unique combination of landscape and other natural features within the study area) explained more variability in contaminant concentrations than did any one or more individual parameters of environmental setting. In general, differences among study areas probably are related to a combination of factors: (1) regional differences in contaminant sources (e.g., atmospheric deposition, pesticide use, dominant industrial activities) that are not reflected in national-scale urban land cover; and (2) study area-specific combinations of basin characteristics (e.g., high precipitation and steep slopes) that affect contaminant transport to, and sediment dilution in, streams. For example, PAH concentrations in Seattle and Salt Lake City are lower (Fig. 2) and have a lower ratio of combustion PAHs/all PAHs (Fig. S5.2) compared with study areas east of the Continental Divide. Recent studies have indicated high PAHs in runoff from coal–tar sealed parking lots (Mahler et al. 2005; Watts et al. 2010), and dust from seal-coated pavement contained 1,000-fold higher PAH concentrations in six cities east of the Continental Divide, where coal–tar-based sealcoat predominates, than in three western cities where asphalt-based sealcoat predominates (Van Metre et al. 2009).

Three study areas are noteworthy because their contaminant concentrations were particularly high (Boston) or low (Seattle) or because they were significant most often in the 12 contaminant models (Seattle and Salt Lake City). Although Boston streams had the highest median concentrations of ten of the 12 contaminants (PAHs, chlordane, DDT, PCBs, arsenic, cadmium, copper, lead, mercury, and zinc; Figs. 2 and 3), the high concentrations observed were consistent with Boston's urbanization and high sediment-TOC for six of these ten contaminants (i.e., the study area indicator variable for Boston was not significant). Models for the other four contaminants—chlordane, arsenic, lead, and mercury—included Boston as a positive study-area variable, thus indicating higher concentrations than expected relative to other sites in the aggregated data set that have comparable urbanization and sediment-TOC. This may reflect the long history of industrial activity and

high density of development in the Boston area or the geochemistry in the region (e.g., high arsenic in association with calcareous metamorphic bedrock; Robinson et al. 2004).

Seattle had the lowest concentrations for all six organic contaminants, cadmium, and lead and the second lowest concentrations for copper and zinc (Figs. 2, 3). Seattle was a significant negative study-area variable in six of the ten models (DDT, chlordane, bifenthrin, cadmium, copper, zinc; Table 2), indicating lower concentrations than other study areas for sites with similar urbanization and sediment-TOC. Reasons are unknown, but possibilities include the high sand content in Seattle sediment samples, which may decrease contaminant sorption (although TOC normalization should decrease this potential); less developed riparian corridors; and shorter retention times (i.e., high precipitation and substantial slopes) in Seattle compared with other study areas.

Salt Lake City was a significant positive variable in seven contaminant models, including bifenthrin, dieldrin, and all trace elements except arsenic (Table 2). Greater-than-expected concentrations of dieldrin and bifenthrin (the only models based on the termite-urban score) may result if this score under-represents urban insecticide use in the area, such as control of ants and scorpions. Greater-than-expected trace element concentrations (cadmium, chromium, copper, lead, nickel, and zinc) may result from historical mining and smelting activities in some basins (Waddell et al. 2004). However, Salt Lake City also is unique in that basin characteristics were determined only for the valley floor portion of the basin to account for noncontributing drainage from upper basins that were subject to water diversion and impoundment (Waddell et al. 2004; Moran et al. 2012). If this procedure underestimated urban contribution to drainage at sampling sites, this may have contributed to greater-than-expected contaminant concentrations.

Comparison with SQGs

One or more PECs for individual contaminants were exceeded in 16 % of sediment samples: arsenic (at one Boston site); individual PAHs (at two Denver sites); bifenthrin (at 14 sites in six study areas [all except Milwaukee]); permethrin (one Denver site); and cyhalothrin and cypermethrin (one site each in Dallas). The ESB for dieldrin was not exceeded in any samples, all of which had dieldrin concentrations less then one tenth of the ESB value.

The $\sum$ESBTU for PAH mixtures was exceeded in 20 sediment samples (20 %) from five study areas as indicated in Fig. 8. Similarly, the summed toxicity quotients for pyrethroids, which are expected to have additive toxicity (Trimble et al. 2009), exceeded the toxicity threshold in 15 sediment samples (15 %), of which 14 samples also exceeded the toxicity threshold for at least one individual pyrethroid compound. Altogether, a total of 33 of 98 samples (34 %) exceeded one or more SQGs for individual contaminants or classes—PECs, pyrethroid toxicity quotients, or $\sum$ESBTU for PAHs—thus, these sites are expected to have a high probability of toxicity to benthic invertebrates. Five samples exceeded two or more individual SQGs.

The overall mean PECQ-5B, mean PECQ-5M, and mean PECQ-5P were each compared with the toxicity threshold of 0.1 to assess the likelihood that a sediment sample mixture would be toxic to sediment-dwelling organisms (Ingersoll et al. 2001; Kemble et al. 2012). The 0.1 threshold for contaminant mixtures was exceeded in 36 % of samples for the mean PECQ-5B compared with 20 % for the mean PECQ-5M and 37 % of samples for the mean PECQ-5P. Combining predictions from the mean PECQ-5B and all individual SQGs, a total of 45 % of samples were identified as potentially toxic (Fig. 8). The efficacy of various SQGs in explaining measured toxicity is described in Kemble et al. (2012).

For the most part, exceedances of individual SQGs and the 0.1 threshold for mean PECQ-5B occurred at sites with >40 % urban land in the basin. There were five exceptions: The Stillwater River in Boston (with 8 % urban land in the basin) exceeded the arsenic PEC. Buffalo Creek in Dallas (23 % urban) and Hoods Creek in Milwaukee (19 % urban) both exceeded the mean PECQ-5B threshold but no other SQGs; these basins contained approximately 30 and 65 % agricultural land, respectively, so agricultural bifenthrin use may have contributed to their bifenthrin toxicity quotients. Bear and Cherry Creeks in Denver (6 and 18 % urban land, respectively) exceeded multiple SQGs; these sites were outliers in several analyses (e.g., Fig. 4d), probably because water from undeveloped upper portions of the basin was often diverted or impounded so that basin-scale land-use data underestimated the urban contribution to water flow at the sampling sites (Sprague et al. 2006).

Several caveats apply to the mean PECQs and pyrethroid sediment toxicity thresholds used in the current article as predictors of toxicity and discussed in relation to observed toxicity in Kemble et al. (2012): (1) the predicted contributions of pyrethroids to the observed sediment toxicity is subject to uncertainty in 28-day amphipod toxicity thresholds estimated for the pyrethroids; (2) the mean PECQ-5 (regardless of which method is used to incorporate pyrethroids) could underestimate the contribution of pyrethroids relative to other contaminants because all three methods assume that nondetections equal zero for pyrethroids but one half of the reporting level for other

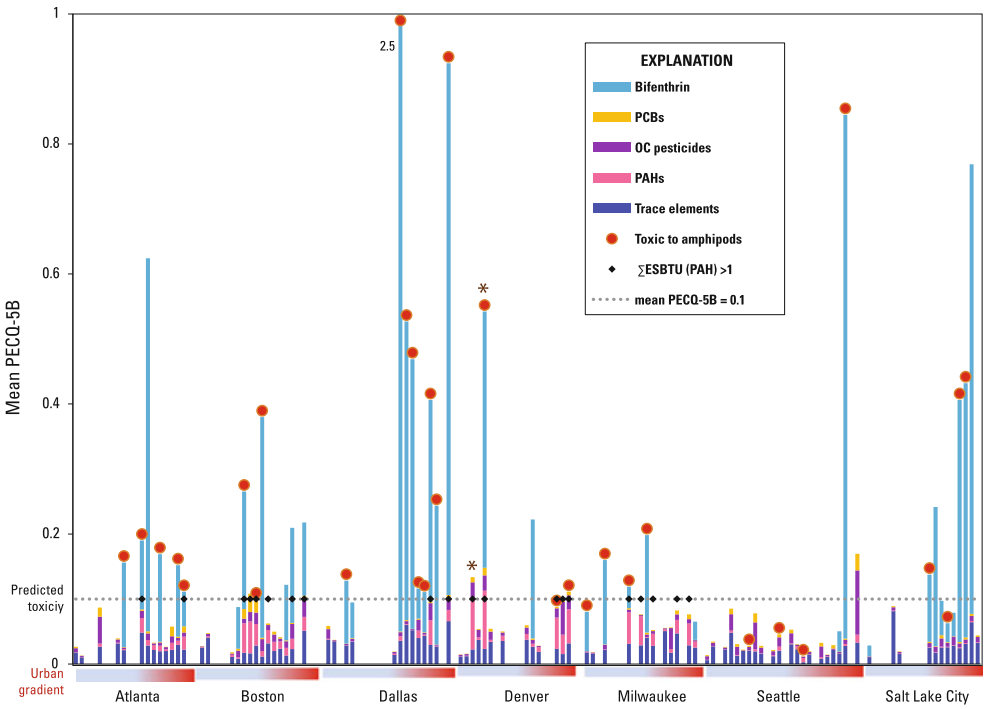


Fig. 8 Summary of toxicity-weighted contaminant concentrations (mean PECQ-5B) and measured sediment toxicity to the amphipod *H. azteca* in 28-day tests (Kemble et al. 2012) at each site along the urban gradient in all seven study areas. Each site has a *stacked bar* showing the contribution of each contaminant class (as color segments) to the mean PECQ-5B. Sites with either a *black diamond* (indicating ∑ESBTU for PAHs >1) or a stacked bar height above the *dotted line* (mean PECQ-5B >0.1) were predicted to be toxic based on these SQGs. Shading from *gray* to *red* below the *x*-axis represents increasing urban land use within the study area. Sites (*stacked bars*) are spaced along the *x*-axis for each study area according to the basin urban land use from 0 to 100 %. *Site for which urban streamflow is higher than represented by basin urban land use because of water diversion and impoundment (Bear and Cherry Creeks)

contaminants; (3) the collection of sediment from depositional areas and sieving of sediments may overestimate both contaminant concentrations and SQG exceedances in stream sediment by removing fractions with lower contaminant levels that would effectively dilute exposure; (4) sediment-associated contaminants that do not exceed thresholds (e.g., resmethrin) or that were not determined in the present study (e.g., dioxins and furans) were not included in mean PECQ calculations but nevertheless may have effects on aquatic biota; and (5) for trace elements, the use of strong acid digestion yielded total concentrations in sediment rather than bioavailable concentrations. The PECs are empirically based, and the studies from which PECs were generated (spiked sediment bioassays and field studies with matching sediment chemistry and biological effects data) varyingly used strong acid and weak acid digestions to extract trace elements (MacDonald 1994; MacDonald et al. 2000), so comparison with these PEC values may overestimate the likelihood of toxicity to trace elements. Bioavailable trace element concentrations are expected to be 30–40 % lower than total concentrations in the sediment matrix (Ingersoll et al. 2009). As noted, some of these factors would either bias the mean PECQ low or high.

Summary

Figure 8 summarizes the overall results from the present study, including sediment contamination and predicted toxicity inferred from SQGs, as described herein, combined with observed toxicity to the amphipod *H. azteca* reported in Kemble et al. (2012). The mean PECQ-5B tended to be higher at sites with >40 % urban land in the basin. However, the considerable variability in mean PECQ-5B values in these urban streams indicates that contaminant concentrations are affected by other factors, both within and among study areas. The response to urbanization differed among metropolitan areas for sediment contamination and toxicity (Fig. 8) as was observed in previous studies for water chemistry and ecological communities (Sprague and Nowell 2008; Brown et al. 2009; Coles et al. 2009, 2012). Differences among study areas in sediment contamination were significant even after accounting for differing levels of urbanization and sediment-TOC. Where pyrethroids were present, they accounted for a large share of the toxicity-weighted contaminant concentration (mean PECQ-5B; also mean PECQ-5M and mean PECQ-5P [not shown in Fig. 8]). Bifenthrin, in particular, is a dramatic example of how a single, relatively new contaminant may

dominate potential toxicity in stream sediment from several metropolitan areas across the United States. Figure 8 also summarizes the incidence of observed toxicity to amphipods in the sampled sediments, as reported in part 2 of this series (Kemble et al. 2012), in relation to degree of urbanization, study area, and the mean PECQ-5B. Kemble et al. (2012) discuss the relations between sediment chemistry and toxicity, including the contribution of pyrethroids and other contaminants associated with mean PECQ, observed in amphipods and the midge. The results of the present study confirm the importance of bifenthrin as a primary cause of potential toxicity in urban streams reported previously for residential creeks, especially near storm drain outfalls, in parts of California (Weston et al. 2005; Amweg et al. 2006; Holmes et al. 2008), Illinois (Ding et al. 2010), and Texas (Hintzen et al. 2009). Findings for the pyrethroids illustrate the importance of tracking new contaminants introduced to aquatic ecosystems and the development of analytical methods and toxicity thresholds to support the assessment and management of contaminated sediments.

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