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Land-use-based sources and trends of dissolved PBDEs and PAHs in an urbanized watershed using passive polyethylene samplers

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Abstract

Narragansett Bay is a temperate estuary on the Atlantic coast of Rhode Island in North United States, which receives organic pollutants from urban and industrial activities in its watershed, though detailed knowledge on sources and fluxes is missing. Twenty-four polyethylene passive samplers were deployed in the surface water of the watershed around Narragansett Bay during June-July of 2014, to examine the spatial variability and possible sources of priority pollutants, namely dissolved polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs). Dissolved $\sum_{22}$PAH concentrations ranged from 3.6 - 340 ng L$^{-1}$, and from 2.9 - 220 pg L$^{-1}$ for $\sum_{12}$PBDE. The spatial variability of the concentrations was correlated to land use pattern and population distribution, in particular with human activities within 2 km of sampling sites. River discharges derived from the concentrations of PAHs and PBDEs measured here were 10-20 times greater than their previously measured concentrations in the open waters of Narragansett Bay. These results imply that river waters are the main source of PAHs and PDBEs to the Bay and that major sink terms (e.g., sedimentation, degradation) affect their concentrations in the estuary. Predicted PAH and PBDE toxicity based on dissolved concentrations did not exceed 1 toxic unit, suggested that no toxicity occurred at the sampling sites.

Capsule: This work represents the first survey of dissolved PAHs and PBDEs using passive samplers in an urbanized estuary’s watershed, helping to constrain pollutant flows from land-based sources.
Keywords: Polycyclic aromatic hydrocarbons (PAHs); Polybrominated diphenyl ethers (PBDEs); polyethylene passive samplers; urbanized watershed; land-use-based source.

Introduction

Estuaries are subjected to multiple anthropogenic or naturally occurring stress factors, and prone to be affected to different degrees by an array of pollutants including hydrophobic organic contaminants (HOCs). Polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs) are two typical classes of HOCs which continue to be released into the environment mainly via natural (PAHs) and anthropogenic processes (PAHs and PDBEs) (Yunker et al., 2002; Hites, 2004; Lima et al., 2005; Takada, 2010; Parinos et al., 2013). In aquatic ecosystems, the dissolved PAHs and PBDEs are subjected to evaporation, adsorption to suspended particulates, uptake by aquatic organisms and bioaccumulation, bio-degradation and photo-degradation (Rayne and Ikonomou, 2005; Tsapakis et al., 2006; Guigue et al., 2014). They can be further transferred into the aquatic environment by urban run-off, municipal sewage, industrial effluents, oil spillage or leakage, gas-water exchange, and atmospheric deposition (Lima et al., 2005; Song et al., 2006; Tsapakis et al., 2006; Parinos et al., 2013). Previous work has suggested that aquatic ecosystems with high PAH and PBDE concentrations might pose potential ecological risks, causing carcinogenic, mutagenic, and toxic effects on aquatic organisms (Hall et al., 2003; Oliveira et al., 2012; Ikenaka et al., 2013).
Narragansett Bay is a temperate estuary on the Atlantic coast of Rhode Island, USA. In previous work, the concentration of PAHs in Narragansett Bay and other coastal waters was shown to have strong correlations with urban runoff, river flows and wastewater treatment plant discharges (Lohmann et al., 2011). Due to its long industrial history, PAHs are present in the sediments of Narragansett Bay (Hoffman et al., 1984; Pruell et al., 1986; Latimer and Quinn, 1996; Lima et al., 2003; Hartmann et al., 2004; Lohmann et al., 2011). Concentrations and profiles of PAHs in the water column of Narragansett Bay were reported (Lohmann et al., 2011; Yonis, 2012), but so far no study has actually measured PAHs in the watershed and contributing rivers directly. Likewise, the freely dissolved PBDEs in Narragansett Bay probably originate from a combination of air-water exchange, freshwater runoff, rivers, and wastewater treatment plants (Sacks and Lohmann, 2012), and are still present in currently products, suggesting on-going releases to Narragansett Bay and the surrounding areas. This prompted us to investigate the relationship between the land-based activities and PAHs and PBDEs concentrations using passive samplers, and whether river inputs responsible for the contaminants in Narragansett Bay.

Low density polyethylene (LDPE) passive samplers are cost-effective, technically simple tools with lower detection limits than traditional active sampling techniques (Mcdonough et al., 2014). Instead of pumping air or water through a filter, passive sampling relies on diffusion to accumulate analytes of interests and only accumulates molecules which are freely dissolved in the water or the atmosphere (Adams et al., 2007). This avoids the analytical interference of, e.g., dissolved organic carbon (DOC)
which can affect active sampling results (Lohmann et al., 2011). Moreover, concentrations of freely dissolved PAHs and PBDEs are of interest because this fraction is available for direct diffusive exchange between water and other reservoirs such as air, biota, or sediment (Kane Driscoll et al., 2010; Mhadhbi et al., 2012; Bragin et al., 2017). Different passive samplers have been used in seawaters, harbors, rivers and urban waterways to determine freely dissolved concentrations of PAHs and PBDEs (Cornelissen et al., 2008a; Sower and Anderson, 2008; Lohmann et al., 2011; Lohmann et al., 2012; Sacks and Lohmann, 2012; Alvarez et al., 2014).

The objectives of this study were to enhance our understanding of the distribution and behavior of PAHs and PBDEs across the Narragansett Bay watershed. We deployed LDPE samplers in the surface water at 24 sites across the watershed in June-July 2014 with the aim of (1) establishing baseline concentrations and spatial trends of dissolved PAHs and PBDEs in the Narragansett Bay watershed, (2) diagnosing the main sources of dissolved PAHs and PBDEs to Narragansett Bay, (3) evaluating the importance of land-based sources of dissolved PAHs and PBDEs by investigating the relationship between land-use characteristics and concentrations, (4) elucidating the main factors controlling the fate of PAHs and PBDEs in the watershed, and (5) lastly predicting and assessing the PAH and PBDE toxicity.

Materials and Methods

Study area

The Narragansett watershed area in Rhode Island was 2077.6 km² including the
estuarine waters (43.6% total watershed). The land coverage pattern around the bay was
dominated by forest (58%), urban or built-up land (28%), agriculture (5.4%), water
(4%), wetland, barren land and rangeland (each < 2%, see TOC).

Preparation and deployment of LDPE passive samplers

Fifty-one micrometer thick LDPE samplers were cut and cleaned for yielding
passive samplers. Deuterated PAHs (pyrene-d10 and benzo(a)pyrene-d12) and
brominated biphenyls (2,5-Dibromobiphenyl, 2,2’,5,5’-Tetrabromobiphenyl,
2,2’,4,5’,6-Pentabromobiphenyl, and octachloronaphthalene) were used as
performance reference compounds (PRCs) to infer the equilibrium concentration of
compounds in the passive samplers as in previous work (Booij et al., 2002; Mcdonough
et al., 2014). Both passive samplers and field blanks were picked up by trained
volunteers and deployed in surface waters throughout the Narragansett Bay watershed
in June-July 2014, as a part of the University of Rhode Island Watershed Watch program
(http://www.uri.edu/ce/wq/ww/ index.htm). The detail methodology, map of
monitoring sites, and other related monitoring summary were provided in the
supporting information (Figure S1 & Table S1).

Analysis

LDPE samplers were wiped clean with Kimwipes and extracted once in hexane
for 24 h after addition of 20 ng of labeled PAHs (acenaphthalene-d10, phenanthrene-
d10, chrysene-d12 and perylene-d12) and PBDEs (\(^{13}\)C\(_{12}\)-BDE28, \(^{13}\)C\(_{12}\)-BDE47, \(^{13}\)C\(_{12}\)-
BDE99, $^{13}$C$_{12}$-BDE153 and $^{13}$C$_{12}$-BDE183) surrogates. Extracted LDPE samplers were air dried and weighed. Extracts were concentrated to 100 μL, and spiked with 50 ng of p-terphenyl-d$_{14}$ (injection standard) directly before instrumental analysis.

PAHs were analyzed using an Agilent 6890 GC System coupled with an Agilent 5973 Network Mass Selective Detector and quantified using Agilent Technologies MSD Productivity ChemStation D.03.00552 software as presented elsewhere (Lohmann et al., 2012). PBDEs were analyzed on a Waters Quattro micro GS Micromass MS-MS and quantified using Waters QuanLynx V4.1 software as detailed elsewhere (Sacks and Lohmann, 2012). Further details are given in the Supporting Information. Samples were analyzed for 22 PAHs and 12 PBDEs (Table S2 and S3).

Quality assurance/Quality control

Method blanks were prepared with each batch of samples to monitor for laboratory contamination. Samples were blank-corrected by subtracting the average of the method and field blank concentrations. Target analytes were quantified by internal calibration relative to surrogates. Average surrogate recoveries for PAHs and PBDEs ranged from 68%-86% and 39%-62%, respectively (Tables S2 to S4). Method detection limits (MDLs) were calculated as 3 times the standard deviation of the average field blank concentration. Concentrations < MDL were reported as half of the MDL, as recommended previously (Antweiler and Taylor, 2008). Compounds that were measured above the MDL in less than 20% of samples were omitted from the discussion.
Calculations/Data analysis

- Determination of sampling rate and ambient concentration

The fraction of equilibrium \( f \) achieved for each compound was determined by fitting the equilibrium of the PRCs and their temperature-corrected log \( K_{PE} \) values to a model curve derived as,

\[
f = 1 - e^{-\frac{R_s t}{K_{PEw} M_{PE}}}\]

Where \( R_s \) (L/day) is the sampling rate defined as the amount of water that comes into contact with the sampler per day; \( t \) is deployment time (days); \( M_{PE} \) is the PE weight (kg); and \( K_{PEw} \) is the LDPE-water partitioning coefficient (L/kg). The values of \( R_s \) can be estimated using nonlinear least squares methods, by considering \( f \) as a continuous function of \( K_{PEw} \), with \( R_s \) as an adjustable parameter using Excel Solver to obtain the best fit (Smedes and Booij, 2010). The average aqueous sampling rate of PAHs and PBDEs was 21 ± 14 L/day and 16 ± 7 L/day, respectively.

Freely dissolved concentrations, \( C_W \) (ng/L) of compounds were calculated from the equation,

\[
C_W = \frac{C_{PE}}{K_{PEw} \left( 1 - e^{-\frac{R_s t}{K_{PEw} M_{PE}}} \right)}
\]

Where \( C_{PE} \) is the PE-normalized concentrations (ng/L). For more details, see the Supporting Information and Tables S1.

- Land-use regression and PAHs source analysis

Land-use regressions models are GIS-based spatial models that relate locations specific data on pollutant concentrations to location-specific source and environment
data using regression (Briggs et al., 1997). The following land-use classes were considered: residential areas, commercial and industrial areas, facilities and institutes, and recreational areas occupied by human activities (Figure S2). Other variables included population density (Pop/km²), buildings, roads, sewer areas, and impervious surface areas. All geospatial data were retrieved from the RIGIS web site (http://www.edc.uri.edu/rigis). The analyses were conducted using ArcGIS version 10.2 software using coverage and grid data formats whenever possible. Statistical analyses were performed using IBM SPSS Statistics 22. Linear regression was chosen as an appropriate model based on the correlation between dissolved concentrations and major descriptor variables. For more details, also for assessing PAHs sources, see the Supporting Information.

Toxicity assessment

A toxic unit (TU) is defined as the ratio of the concentration in a medium to the effect concentration in that medium (USEPA, 2003). The TUs for EPA-34 individual PAHs, \( C_{\text{pw, EPA-34}} \) (TUs), were considered one of the most accurate measures for evaluating the toxicity of pyrogenic PAH-impacted sites (Arp et al., 2011). The toxic/nontoxic threshold value for the protection of sensitive benthic organisms is 1.0 TU (USEPA, 2003). In this study, we chose pyrene, fluoranthene, benz(a)anthracene, and chrysene, to estimate the \( C_{\text{pw, EPA-34}} \) (TUs) and to predict the potential toxicity of each sampling site by using the models in Arp et al. (2011), since these compounds were the most commonly quantified compound in surface water (96-100% of samples, see in
Table S2) and also exhibited good correlations with the other PAHs.

For PBDEs, the risk assessment for water organisms was performed on the basis of the risk quotients (RQs) for non target organisms (Santos et al., 2007; Sánchez-Avila et al., 2012). RQs were calculated as a quotient of the measured environmental concentration (MEC) and the predicted no effect concentration (PNEC), and were characterized using the maximum probable risks for ecological effect guideline established by Marcus et al. (2010) as: no significant risk (RQ<1.0), a small potential for adverse effects (1.0 ≤ RQ<10), significant potential for adverse effects (10 ≤ RQ<100), and adverse effects should be expected (RQ≥100). The risk assessment was performed considering three trophic levels of fish, Daphnia and algae, following the recommendation of the Technical Guidance Document on Risk Assessment (European Commission, 2003). For more details, see the Supporting Information.

**Results and Discussion**

**Dissolved PAHs & PBDEs in surface water**

- Concentrations and spatial trends

The concentrations of dissolved $\sum_{22}$PAH in the surface water ranged from 3.6 to 340 ng L$^{-1}$ with the mean value of 44 ng L$^{-1}$ (Figure 1, Table S5). The dominant PAHs were phenanthrene (5-53%), pyrene (5-43%) and fluoranthene (<MDL-38%). Phenanthrene contributed more to the dissolved $\sum_{22}$PAH in the lower watershed, while pyrene and fluoranthene were the main contributors in the upper areas. Retene accounted for 2.9-19.4% of the total freely dissolved concentrations.
PAH concentrations were greater in the upper, more urban/industrialized watershed, and in coastal areas. The greatest dissolved $\Sigma_{22}$PAH concentration was recorded at Knowles Brook (BB_KB), followed by Woonasquatucket River near Donigian Park (WR_DP) (184 ng L$^{-1}$), and Mill Brook near Greenwich Bay (GB4) (75 ng L$^{-1}$). Sites around Bristol Harbor also had relatively high concentrations (BH8-10 ng L$^{-1}$, BH1-57 ng L$^{-1}$, BH12-18 ng L$^{-1}$). These sites were, by and large, also surrounded by largest population densities.

Freely dissolved concentrations of $\Sigma_{12}$PBDE in the surface water were more than three orders of magnitude lower than PAHs across the Narragansett Bay watershed. PBDE concentrations ranged from 2.9 to 220 pg L$^{-1}$ with a mean value of 25 pg L$^{-1}$ (Figure 2, Table S6). Dissolved PBDEs were dominated by BDE-47 (29-66%), BDE-99 (16-40%) and BDE-100 (2-14%). PBDE concentrations were also greater in the upper watershed and coastal areas. Dissolved $\Sigma_{12}$PBDE concentrations were greatest in rivers (Woonasquatucket River, at 224 pg L$^{-1}$, followed by the Pawtuxet River, at 150 pg L$^{-1}$).

The presence of PAHs in the surface water, away from major urban/industrialized sources, most likely stemmed from atmospheric deposition, and represented background contamination of PAHs across the northeastern United States. These were typically characterized by low concentrations of PAHs, typically below 10 ng L$^{-1}$, as can be seen for all samplers deployed in the southern half of the state. All northern sites, with the exception of an upstream river sample displayed much greater PAH concentrations, ranging from 10 to greater than 300 ng L$^{-1}$. 
For PBDEs, although the gradient from north to south was not as strong as for PAHs, a similar separation of sites could be seen. The lowest concentrations of PBDEs occurred in the southern samples, which ranged from 3-19 pg L$^{-1}$, while the northern samples displayed $\sum$PBDEs from 4.4 up to 224 pg L$^{-1}$.

Comparison to other studies

PAH concentrations derived here were about 3-9 times higher than those obtained by passive sampling in the Great Lakes (USA/Canada) (Ruge, 2013; McDonough et al., 2014), and about 5 times higher than those reported in the seawater of Narragansett Bay (Table S7) (Yonis, 2012). Clearly, surface waters continued to act as sources of PAHs to the Narragansett Bay estuary and the ocean. PAH concentrations were comparable to those of southern Chesapeake Bay in the USA, the River Seine and its estuary in France, and on the Dalian coast in China (Gustafson and Dickhut, 1997; Fernandes et al., 1997; Liu et al., 2013).

The concentrations of PBDEs in the present study were more than 20 times higher than those also measured with PE samplers in the seawater of Narragansett Bay, off Dutch Island, and those in the North American Great Lakes (Table S7) (Booij et al., 2002; Sacks and Lohmann, 2012; Ruge, 2013). Also for PDBEs, surface waters continue to act as sources to the estuary and the ocean. The dissolved PBDEs in our study area were similar to those of San Francisco Bay, USA (Latimer and Quinn, 1996) and the NW Mediterranean Sea off Spain (Sánchez-Avila et al., 2012). However, our freely dissolved PBDE values were much lower than those found in the New York/New
Jersey Harbor, Raritan Bay, USA (Zarnadze and Rodenburg, 2008), and Gwangyang Bay, Korea (Kim et al., 2012) (Table S7), though different sampling approaches were used. Active sampling methods used in the compared studies would overestimate and report much higher “dissolved” concentrations of PBDEs than what we found using LDPE samplers in Narragansett Bay (Oros et al., 2005; Sacks and Lohmann, 2012).

Sources of dissolved PAHs

We used established source diagnostics, including diagnostic ratios, PAH profile, principal component analysis to identify where the PAHs originated from. In brief, dissolved PAHs were primarily from pyrogenic sources including fossil fuel combustion at most sites. Some coastal sites were affected by petrogenic sources, and some remote sites by wood combustion. Principal component analysis of dissolved PAHs (Figure S4a) and gaseous PAHs from a previous study (Figure S4b) demonstrated the importance of atmospheric deposition for PAHs. The results of PAH profiles further indicated the deposition and transportation of gasoline vehicle particulate emissions and tire particles as indicators of potential anthropogenic sources (Figure S6 & S7). For more details, see the Supporting Information.

Sources of dissolved PBDEs

Tetra- and penta-BDEs were most commonly detected at the Narragansett Bay watershed sites, likely stemming from the penta-BDE commercial mixture. The contributions of the lower brominated BDEs to the Narragansett Bay watershed
suggested that residences and municipal sewage treatment plant effluents may be significant sources of PBDEs (Hale et al., 2006; Song et al., 2006).

In contrast to PAHs, the results for PDBEs showed that neighboring sites were similar to each other and clustered together (Figure S5). This implies that environmental sources of PBDEs are regional in nature rather than location-specific, and related to the usage of flame retardants in anthropogenic activities, and the proximity to sources.

Land-Use Regression Analysis

Recently, land-use regression methods have been developed and applied in exploring the relations between ambient PAHs and anthropogenic factors (Noth et al., 2011; Melymuk et al., 2013; Machado et al., 2014; Huang et al., 2017; Yao et al., 2017; Zhang et al., 2017). Most researches focus on the PAHs in atmosphere and sediments while only few focus on the dissolved PAHs.

The results for the \( \Sigma \) PAH and each individual compound are given in Table 1, which was able to explain 39-80% of PAH variability. Roads and commercial and industrial areas within 2 km radius explained 73% of the variability in \( \Sigma \)PAH concentrations (Table 1, FigureS8-S10). Road vehicle emissions are credited as a major source of dissolved MMW PAHs in the watershed areas (Figure S8), which were consistent with the results of PAH profiles from gasoline vehicle particulate emissions and tire particles in roads. Population density and sewer areas within 2 km radius were the other variables accounting for \(~40%\) of the variability in LMW PAH concentrations (Acy, Ace, and Flr) (Table 1). For PBDEs, we did not conduct any further regression
analysis since there was weak correlation between the compound and land-use variables
(Table S8, Figure S11).

In recent work, Yang et al (2018), showed that traffic congestion index and the
number of industrial sources within 25-km buffer were the significant predictors of
PAH concentrations in urban lake sediments. Similarly, we find that: 1) transportation
and industry continue to be dominant sources of PAHs to the environment; 2)
monitoring sites should be situated in the relevant buffer zone where PAHs in the
environment are most related to human activities.

PAHs & PBDEs toxicity predictions and assessments

Maybe not surprisingly, no risk for potential effects to aquatic organisms
associated to PBDE was observed, considering short-term exposure of acute toxicity.
ΣRQ were all well below 1, ranging from 1.4 e^{-4} to 5.9 e^{-3}, 4.0 e^{-5} to 3.3e^{-4}, and 4.3 e^{-4}
to 3.4 e^{-2} for fish, algae, and Daphnia magna, respectively (Table S10). Even so, the
continuous intake of PBDEs by water organisms can be accumulated in the food chain.
Therefore, more studies are needed concerning bioaccumulation and biomagnification
of PBDEs in biota for a better risk evaluation of these chemicals in the water
environment.

For PAHs, none of our sites exceeded a TU of one, suggesting that no toxicity
occurred at all the sampling sites (Figure 3). We recognize that our samples represented
surface water, which is generally expected to be less contaminated than pore water. A
few of the highest contaminated sites (Woonasquatucket River, Buckeye Brook and
Greenwich Bay), however, displayed TU values between 0.3-0.7, indicating cause for concern. A consistent trend appeared for $C_{pw, EPA-34}$ (TU) values predicted by different PAHs, which might indicate the potential feasibility of the models using the freely dissolved concentrations from surface water in Narragansett watershed area (Figure 3). This trend further implied the similar pattern or content of PAHs could be found between sediment pore water and surface water due to the river transportation, thus aqueous distribution of PAHs at surface water layer might be sufficient to reflect the overall contamination level for the bulk overlying water column in the watershed (Cornelissen et al., 2008b; Lai et al., 2015). Although, more research should be conducted to prove the above assumption.

Riverine runoff as a vector for transporting PAHs and PDBEs to Narragansett Bay

This study enable us to perform a first estimation of the potential importance of riverine discharges to pollutant concentrations in the Narragansett Bay estuary itself (Figure S12). At steady-state, river water inflow concentrations would be diluted by ca 12 times in the estuary (ratio of freshwater flow times flushing time divided by the total volume) (Pilson, 1985). The Pawtuxet and Woonasquatucket Rivers were measured near their discharge into the Bay; sum PAHs ranged from 70-180 ng L$^{-1}$, while sum PBDEs were 150-220 ng L$^{-1}$. Previously measured PAH concentrations, unaffected by major storms, ranged from 0.5-2 ng L$^{-1}$ for open water stations (Lohmann et al., 2011). Dissolved PAH concentrations in the estuary are lower, by almost an order of magnitude, to what can be expected based on the dilution of riverine discharges (6-15 ng L$^{-1}$). For sum
PBDEs, previously measured concentrations ranged from 6.5 pg L$^{-1}$ in the northern part of the Bay to below detection limit in the southern part (Sacks and Lohmann, 2012). Overall, the open water concentrations for Narragansett Bay are again ~10-times lower than expected from the dilution of riverine inflows (12-20 pg L$^{-1}$). Common processes that could reduce dissolved concentrations of both PAHs and PDBEs in the estuary include sorption to solids, sedimentation, and degradation.

**Implications**

This work represents the first survey of dissolved PAHs and PBDEs using passive samplers, deployed by a long-running volunteer network around the Narragansett Bay watershed. Results suggest that this sampling approach is viable and could be extended to get a better grasp on pollutant flows from the terrestrial sources into the Narragansett Bay estuary. In particular, it could be extended to cover temporal trends of concentrations in the watershed, and target all major point sources into the estuary repeatedly. Surprisingly, several sites appear close to toxic effects based on the presence of key PAHs. The results presented here suggest that rivers are the main conduits of PAHs and PDBEs into the estuary, and imply that neither air-water exchange nor particle-bound deposition of pollutants affects dissolved concentrations to a significant degree. Instead, the evidence indicates that major sinks exists for both PAHs and PBDEs upon their discharge into the estuary, probably including uptake by sediment and degradation. These results should be tested in future work.
Supporting Information

Additional details relating to sampling locations, chemical analysis, and dissolved concentrations of PAHs and PBDEs are available free of charge via the Internet at http://pubs.acs.org.

Acknowledgments

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Figure 1. Dissolved $\sum_{22}$PAH and individual PAHs in the Narragansett Bay watershed.
Figure 2. Dissolved $\sum_{12}$PBDE and individual PBDE in the Narragansett Bay watershed
Figure 3. Estimating the total EPA-34 freely dissolved pore water concentration, $C_{pw, EPA-34}$, expressed as toxic units (TUs) from the freely dissolved concentrations from surface water in Narragansett watershed.
Table 1. Multiple Linear Regression Results for PAHs

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<th>Compound</th>
<th>Variable</th>
<th>ln(A+1)</th>
<th>ln(B+1)</th>
<th>ln(C+1)</th>
<th>ln(D+1)</th>
<th>ln((\Sigma) PAH+1)</th>
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<th>0.353</th>
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<th>/</th>
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<td>/</td>
<td>/</td>
<td>/</td>
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<td>ln(Flr+1)</td>
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<td>/</td>
<td>/</td>
<td>/</td>
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</table>

A = Roads within 2 km;
B = Commercial and industrial areas within 2 km;
C = Population density within 2 km;
D = Sewer area within 2 km;

The p values for these multiple regression equations were less or equal to 0.001.