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Identification of Stormwater Pollution Hotspots in Charleston Peninsula

Ashleigh Kirker[1,2] and Vijay M. Vulava[1]

AUTHORS: [1]College of Charleston, Department of Geology and Environmental Geosciences, 66 George St., Charleston, SC 29466, USA. [2]Temple University, Department of Earth and Environmental Sciences, 1901 North 13th Street, Philadelphia, PA, USA.

Abstract. Flooding is of great concern in fast-growing coastal communities, especially in the southeastern US, due to multiplying threats such as extreme precipitation, coastal storms, and rising sea levels. Contamination associated with stormwater runoff is often given less attention during stormwater planning and management decisions. The US EPA has long recognized that stormwater runoff is the biggest contributor to the impairment of water bodies in the US. In this study, we studied stormwater runoff contamination in a densely developed section of downtown Charleston, South Carolina, to better understand the extent of the problem and identify potential hotspots that could aid in future stormwater management decisions. We focused on a 4.25 km² section of peninsular Charleston that has a dense mix of residential, commercial, and industrial land use. High-resolution 2.5-m elevation data was used to subdivide the research area resulting in four distinctive subwatersheds, each of which had a distinctive land-use pattern. For 16 months starting in September 2016, stormwater samples were collected near storm drains at 24 sites distributed within the 4 subwatersheds immediately after large rainfall events. These water samples were analyzed for enterococci (a fecal indicator bacteria), trace metals (As, Cd, Cr, Cu, Pb, Ni, V, and Zn), and nutrient (NO_3^- and PO_4^{3-}) concentrations. Our data indicated that enterococci concentrations were extremely high in the entire watershed and that these concentrations tended to be higher on days where there was antecedent rain preceding the sample collection. These concentrations were also higher during warmer times of the year (July–September). Trace metals were detected in all filtered water samples, and these concentrations positively correlated with traffic patterns and hence were more prevalent in areas of high traffic. Nutrient ions were present in all water samples, while the PO_4^{3-} concentrations exceeded US EPA ecological standards; PO_4^{3-} concentrations were highest in the subwatershed with the highest residential land use. By coupling these stormwater quality data to watershed delineation, weather conditions, and land-use patterns, we were able to identify general hotspots for stormwater contaminants. The data suggest that there would be public health concerns in areas that are disproportionately affected by stormwater flooding. These insights into the myriad ways natural water systems in fragile coastal ecosystems are being impaired can be employed in stormwater management. We recommend that government agencies include stormwater quality concerns in future planning.

INTRODUCTION

The US Census Bureau reported that between the years 1960 and 2008, the US population grew fastest along the Atlantic, the Pacific, and the Gulf of Mexico shorelines compared with the rest of the country (Wilson and Fischetti 2010). In the most recent decade (2010–2019), population growth was higher in nearly all of South Carolina's coastal counties when compared with overall South Carolina (https://www.census.gov/quickfacts/fact/table/US/PST045219). Similar higher population growth has been reported (at least 17.5% growth in the most recent decade compared with 6.3% growth across the US) in the densely populated coastal counties of Berkeley, Charleston, and Dorchester. These counties currently have a significantly higher population density compared with the state of South Carolina and a much higher urban footprint as well. The coastal watershed in this region, which includes the City of Charleston, spans Berkeley, Charleston, and Dorchester (BCD) counties and is part of the Santee River Basin (Hughes et al. 2000). Recent forecasts predicted that urbanization around Charleston, South Carolina, will triple by 2030, as the most common form of land-use change is caused by urban expansion (Allen and Lu 2003; Drummond et al. 2015). The US National Climate Assessment indicates that extreme precipitation along with rapid sea-level rise will have a significant impact on coastal South Carolina over the next several decades (NOAA 2017). Water quality in the coastal water of this region is also expected to severely degrade as a result of this growth (Allen and Lu 2003). Increased impervious surfaces increase stormwater runoff volume and are linked to habitat degradation from channel erosion and higher pollutant loads (Aryal et al. 2010; Beckingham et al. 2019; Exum et al. 2005). Nonpoint source pollution associated with stormwater runoff is already the most significant cause of surface water impairment in the US (Exum et al. 2005). The most common pollutants include trace metals, polycyclic aromatic hydrocarbons, pathogens, and nutrients (Aryal et al. 2010; Exum et al. 2005).

Microorganisms that are commonly associated with the gut of animals, such as enterococci and *Escherichia coli* (or *E. coli*), are commonly used as indicators for the presence of fecal pathogens in natural water bodies and runoff and thus are referred to as fecal indicator bacteria (FIB) (Selvakumar and Borst 2004). Failing sewage systems, or pet and wild animal waste, are major contributors to the concentration of FIB in stormwater runoff. There is a significant positive correlation between the presence of FIB and urbanization of land upstream of an open water body when compared to undeveloped land (Van Dolah et al. 2008).

Trace metals are commonly present in the urban environment and are especially concentrated in urban/industrial areas due to brake and tire wear, vehicle exhaust, and industrial activities (Aryal et al. 2010). Trace metals such as As may be present because of natural sources such as weathering of phosphate rocks (Sanger et al. 1999). Trace metals often accumulate in road dust either directly or as a result of atmospheric deposition during dry periods and either dissolve in runoff or are sorbed to suspended sediments (Ma et al. 2016). Nutrient contamination is also widely present in urban watershed runoff; in particular, nitrogen and phosphorus in the form of NO_3^- and PO_4^3- contributes to the eutrophication of water bodies. There are additional sources of contamination in use and human/animal waste (Aryal et al. 2010).

Stormwater in the coastal urban watershed ultimately discharges into the estuaries causing degradation of coastal water quality. The Charleston Harbor estuary, which includes the Ashley, Cooper, and Wando Rivers, is considered dissolved oxygen-impaired by the US EPA and the SCDHEC (Cantrell 2013). Other studies in the region confirm impairment in other forms as well, including benthic sediment (Sanger et al. 1999), estuarine habitat (Van Dolah et al. 2008), and shellfish, fish, and mammals (Baechler et al. 2020; Fair et al. 2019; Houde et al. 2005). Stormwater runoff has the most significant impact on all coastal environments but is extremely hard to manage due to the diffuse nature of the pollution.

The main goal of this study was to analyze stormwater quality and identify stormwater contamination hotspots in an urban watershed. The study area is the highly developed urban watershed in the historic downtown section of the city of Charleston, South Carolina. Based on the literature review and our preliminary studies, we hypothesized that the stormwater runoff in the city will be contaminated and will reflect the predominant land-use characteristic of a given section of the watershed. To test this hypothesis, we collected stormwater samples in a broad section of Charleston peninsula, which we subdivided into four sections based on the predominant flow direction of the stormwater runoff. In each of these sections (subwatersheds), we collected discrete stormwater samples during significant rain events that generated sheetflow and runoff between September 2016 and January 2018. By combining water quality with the spatial and statistical analysis, we determined significant hotspots for different sets of contaminants and potential sources of contamination. This approach can be useful in understanding the factors involved in urban stormwater contamination as well as in its subsequent management. The general approach or framework can be adapted to other settings.

MATERIALS AND METHOD

SITE DESCRIPTION

The City of Charleston, South Carolina, is located within the Southeastern Atlantic Lower Coastal Plain (Figure 1). The land area is approximately 290 km², of which the historic peninsula makes up approximately 21 km². The natural, unaltered watershed in this region is forested and characterized by a low topographical gradient and shallow water table (Griffin et al. 2014). The Charleston peninsula has undergone significant land-use change since its founding, and since then many changes were made to the natural depressions, wetlands, and salt marshes by draining and/or filling these areas (Butler 2020). In the decades since 1970, rapid population growth in the region has resulted in an acceleration of land-use change across the region (Allen and Lu 2003; Beckingham et al. 2019).

The average temperature in this region ranges from 9.89 °C in the winter to 28.2 °C in the summer, and the average annual precipitation is approximately 1128 mm yr^{-1} (https://www.weather.gov/chs/climate.) This area receives approximately 41% of its rain during the summer months, which includes a high number of thunderstorms or short, intense storms that contribute to spikes in surface runoff (BCDCOG 2011). More recently, fair weather or sunny day flooding caused by King Tides and rising sea levels have occurred with greater regularity and frequency, causing additional pollution loading and discharges into estuarine waterways (Harris and Ellis 2021; Román-Rivera and Ellis 2018). The Charleston peninsula (Figure 1) is part of the South Carolina Department of Health and Environmental Control's (SCDHEC's) Cooper River Basin (includes EPA

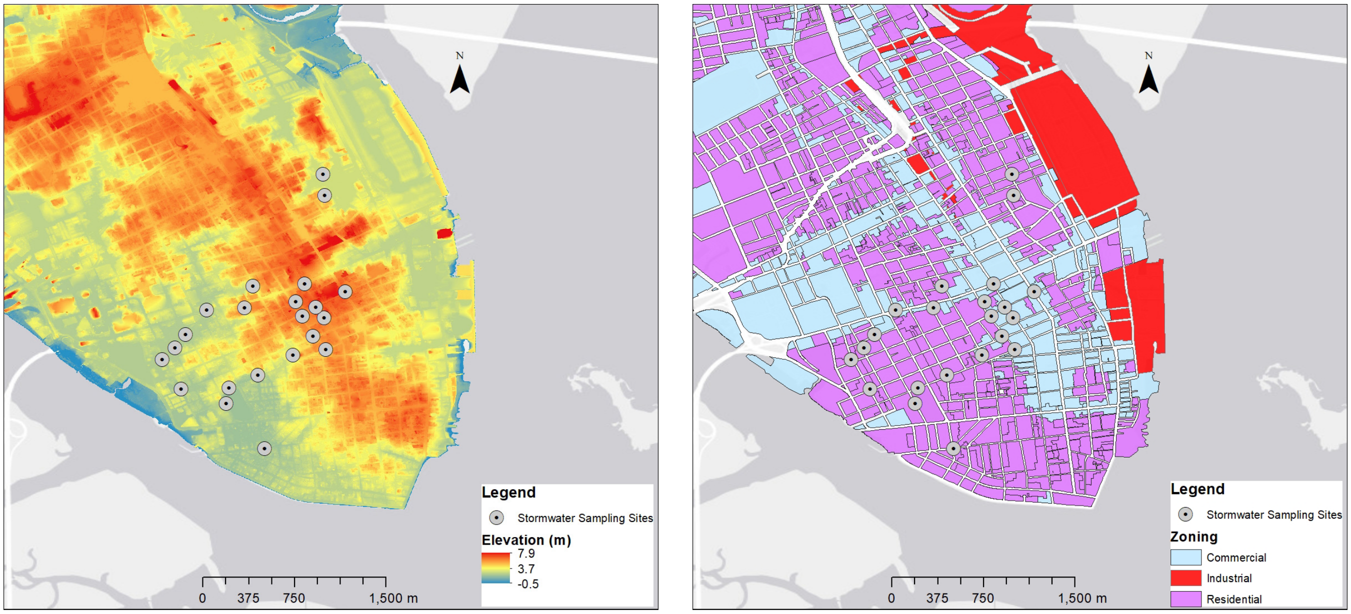

Figure 1. Elevation (left) and land-use (right) patterns of the Charleston peninsula. Stormwater sampling locations are also shown. Sites were chosen at accessible storm drains. Not all sites were sampled during every rain event. Map data sources: USGS, SC DNR, and the city of Charleston

hydrologic units 03050201 and 03050202) and includes parts of the Charleston Harbor and the Cooper, Ashley, and Wando Rivers.

In relatively unaltered environments of this region, the surface soils ranging from sandy-to-loamy types and the subsurface soils ranging from loamy-to-clayey types locally influence natural infiltration and runoff patterns (Griffin et al. 2014). There is very minimal overland flow following rainfall—rainfall usually infiltrates the ground surface, causing the water table to rise and thereby increasing contribution to the baseflow component of stream discharge (Griffin et al. 2014). Natural drainage occurs in broad areas of swamps, wetlands, and tidal marshes. The system is dominated by high tidal amplitudes; because of its low elevation, the broad region is considered estuarine (Houde et al. 2005; Van Dolah et al. 2008). The soils on the Charleston peninsula are classified as UR (Urban land-Yauhannah-Yemassee-Ogeechee association) or urban soil by the USDA-NRCS (http://websoilsurvey.nrcs.usda.gov/). These soils include fill material and have indeterminate soil physical and chemical properties. Because of the high amount of impervious surfaces, the land on the Charleston peninsula has a higher amount of surface runoff compared with unaltered environments (Blair et al. 2014)

STORMWATER SAMPLING

Stormwater samples were collected from an area of approximately 4.25 km^2 of an urban downtown area of the city of Charleston. This area was subdivided into four subwatersheds (Calhoun, Harbor, Colonial, Tradd; see Figure 1) based on watershed delineation, as described in the next section. Between September 2016 and July 2017, 10 rain events were monitored, and stormwater grab samples were collected from multiple sites. For each of the 4 subwatersheds, we canvassed and identified a minimum of 4 sampling sites (Figure 1). The site locations were local topographic low points, where significant stormwater flow into curbside storm drains was observed. In total, 23 sites were sampled during 10 rain events (which are defined as precipitation heavy enough to generate runoff—approximately 1 cm), although not every site was sampled during every rain event. Precipitation data were obtained from NOAA's National Weather Service (NWS) website for downtown Charleston (https://www.weather.gov/chs/climate). The data included the cumulative 3-day precipitation period before the sampling day (antecedent precipitation), as well as the cumulative 24-hour period on the sampling day.

Sampling procedures are adapted from the US EPA standard methods (US EPA 2009). In all cases, stormwater runoff depth near curbside storm drains was deep enough that grab sampling was feasible. Grab samples were collected directly into clean and sterile sample containers, carefully avoiding contact between the road and the sample container without disturbing the sediment at bottom of the water column. Two types of grab samples were collected: (1) samples for fecal indicator bacteria (FIB) analyses and (2) samples for chemical analyses. The first type of samples was collected in 120 mL sterile bottles containing sodium thiosulfate preservative

(IDEXX Laboratories Inc.). These samples were immediately stored in a plastic cooler and prepared for FIB analysis within 6 hours of sampling, as described in the following section. The second type of samples was collected in 50 mL clean, sterile plastic centrifuge tubes (FisherBrand, Thermo Fisher Scientific Inc.) and prepped for chemical analyses, as described in the following section. In all cases, samples and bottles were handled with appropriate personal protective equipment. When needed, water samples were filtered using 0.22 μm polyethersulfone (PES) syringe filters (Millex-GP, Millipore Sigma, Burlington, MA) or diluted using sterile or nonsterile 18.2 MΩ.cm resistivity deionized water.

DELINEATING SUBWATERSHEDS

To identify predominant sheetflow and natural drainage patterns, approximately 4.25 km^2 (Figure 1) of peninsular Charleston was divided into subwatersheds. Preliminary data were obtained from the city of Charleston's Master Floodplain Analysis (Davis & Floyd Inc. 1984) and were coupled with a 2.5-m resolution elevation (Digital Elevation Model, or DEM) lidar dataset (from the South Carolina Department of Natural Resources, http://www.dnr.sc.gov/GIS/lidar.html.) Note that bald earth corrections were not applied to the lidar data to allow the human infrastructure (e.g., building structures) to influence stormwater drainage. The delineation of watersheds used ground surface elevation data to identify the boundary (watershed divide) of an upslope area that contributed to a concentrated outlet or a drain. Typically, contour maps can be used to determine the watershed boundaries (NRCS 1991); however, this method is not very reliable in low-gradient watersheds. In this study, we used the built-in Hydrology toolset of ArcGIS software (ArcGIS Desktop, Esri) for basin delineation (Moore et al. 1991). The 2.5-m resolution DEM data within the area of interest was broken into small grids (2.5 m × 2.5 m) or "cells" whose elevation is known. The Hydrology toolset assumes that there is water present in all cells and identifies the flow direction of water between adjoining cells using the following constraints: (1) flow occurs from higher to lower elevations; (2) when multiple adjacent cells have elevation gradients, flow occurs preferentially toward cells that have steeper gradients; (3) flow accumulates in any cell as water flows from a higher to a lower elevation cell, and (4) flow only occurs when there is a difference in elevations or flow does not occur. The flow direction and accumulation direction identify the streams (and stream orders) that form within a basin, while the no-flow areas help identify the basin boundary. Once the ArcGIS-Hydrology toolset finished the analysis, the locations where water was likely to exit the sample area were identified by analyzing the connected flow paths in the flow direction. Subwatersheds were then delineated with the flow direction raster, using known outfall locations from the city of Charleston's published stormwater sewer network (https://data-charleston-sc.opendata.arcgis.com/) as pour points (outlets). Basin boundaries generally follow high-elevation ridgelines. The Hydrology toolset does not include storm sewers and does not accurately represent subsurface drainage and urban flow networks, so our subwatersheds reflect only overland flow in the study area.

WATER ANALYSIS

To quantify FIB concentrations in water, enterococci bacteria were measured using a standard fluorogenic substrate enterococcus test (Enterolert, IDEXX Laboratories Inc.) (APHA-AWWA-WEF 2017; ASTM 2019). Stormwater samples collected in sterile bottles were diluted 100 times using sterilized deionized water (18 MΩ.cm). Then, a nutrient indicator reagent is added to the sample, mixed thoroughly, and poured into a 96-well Quanti-Tray/2000 (IDEXX) tray, thermally sealed, and incubated for 24 h at 41.0±0.5 °C. All wells that are positive for enterococci bacteria fluoresce under UV light and are quantified using a most probable number (MPN) table to obtain an MPN for each sample. The dilution factors were applied to the final MPN values and were expressed as MPN per 100 mL of stormwater. Both positive controls (*E. faecalis*) and blank samples were incorporated during each week's analyses. These analyses were performed in an SCDHEC-certified lab and were overseen by the lab director and staff.

Dissolved trace metals (As, Cd, Cr, Cu, Pb, Ni, V, and Zn) in water were analyzed using an inductively coupled plasma mass spectrometer (ICP-MS, Agilent 7500cx). All stormwater samples were filtered as described previously and acidified to 2% v/v acidity using HNO_3 (Optima grade, Thermo Fisher Scientific, Inc.). A multi-element standard mix (High Purity Standards) was used to calibrate the ICP-MS. All samples and standards were spiked with 1 μg L^{-1} of Rh and Au internal standards. To account for instrument bias, the mass count ratios of each analyte and an appropriate internal standard were used for quantification. Check standards and blanks (2% v/v HNO^3 in deionized water) were incorporated during analyses of each batch of samples. The linear analytical range for all elements was 10^{-4}-10 mg L^{-1} and the method detection limit was lower than 10^{-4}-10 mg L^{-1}. In all cases, triplicate measurements for each element were less than 5% relative standard deviation (RSD.)

An ion chromatograph (IC, Thermo Dionex ICS-5000+, Thermo Fisher Scientific, Inc.) with a conductivity detector, a microbore isocratic pump, and an electrolytic suppressor was used to measure NO_3^- and PO_4^{3-} concentrations in water samples. An anion exchange column (Thermo IonPac AS22 2×250 mm) paired with 2 guard columns (Thermo IonPac AG22 2×50 mm and Thermo IonPac NG1 2×50 mm) and a 4.5 mM sodium carbonate and 2.0 mM sodium bicarbonate eluent prepared using deionized water (18 MΩ.cm) was used for the ion separations. A 50-μL sample was injected and separated at 0.4 mL min^{-1} for a total elution time of 12 min. A multi-anion standards mix (High Purity Standards) was used to calibrate the peak areas. Laboratory blanks (deionized water) and check standards were incorporated in each batch of samples. A linear analytical detection range of 1-50 mg L^{-1} was obtained with a ±5% RSD for the check standards. Duplicate measurements for samples yielded concentrations within a 5% range, indicating stability of the instrument and the peak integration routines.

STATISTICAL ANALYSES

Due to the large number of analytical variables (dimensions or correlated variables) within the study (e.g., sites, solute types, concentrations, precipitation, locations, sample size), we used principal component analysis (PCA) to reduce the large set of dimensions into a smaller number of dimensions that collectively explain most of the variability in the original set (Christophersen and Hooper 1992; Hair et al. 1998). This method is especially useful in identifying relationships between different variables. An $n \times p$ data matrix (where n is the number of observations and p is the type of observation or the dimensions) was reduced into a lower dimension or principal component space while capturing a good representation of all variability. The first principal component (PC1) is a normalized linear combination of the observations that has the largest variance. Subsequent principal components (PC2, etc.) are normalized linear combinations of observations that are uncorrelated with previous principal components (PC1, etc.) The general expectation was that the first few principal components will account for substantial variation within the data. PCA biplots between PC1 and PC2 were used to project all data as coordinate points, and each type of observation was plotted as a vector pointing toward the direction that represents the maximum correlation between the variable and the principal components. All raw data was scaled so that each of the variables had a mean of 0 and a standard deviation (variance) of 1. A covariance matrix was created for the scaled variables, followed by the calculation of eigenvalues of the covariance matrix. The eigenvector that corresponds to the largest eigenvalue is PC1, and so on. Strong correlations were depicted by the length of the vector. Vectors that were oriented in the same direction (acute angles) indicated that observations were correlated, while inversely correlated variables were oriented in opposite directions (or obtuse angles). Open-source software R (https://www.r-project.org) was used for all statistical computations.

RESULTS

WATERSHED DELINEATION

The four subwatersheds identified were named for the major streets or historical landmarks within each subwatershed (Figure 2). Within each subwatershed, runoff drains into a unique area: the Charleston Marina, the mouth of the Ashley River (seaward of the marina), the Cooper River, or Colonial Lake. The corresponding subwatersheds are

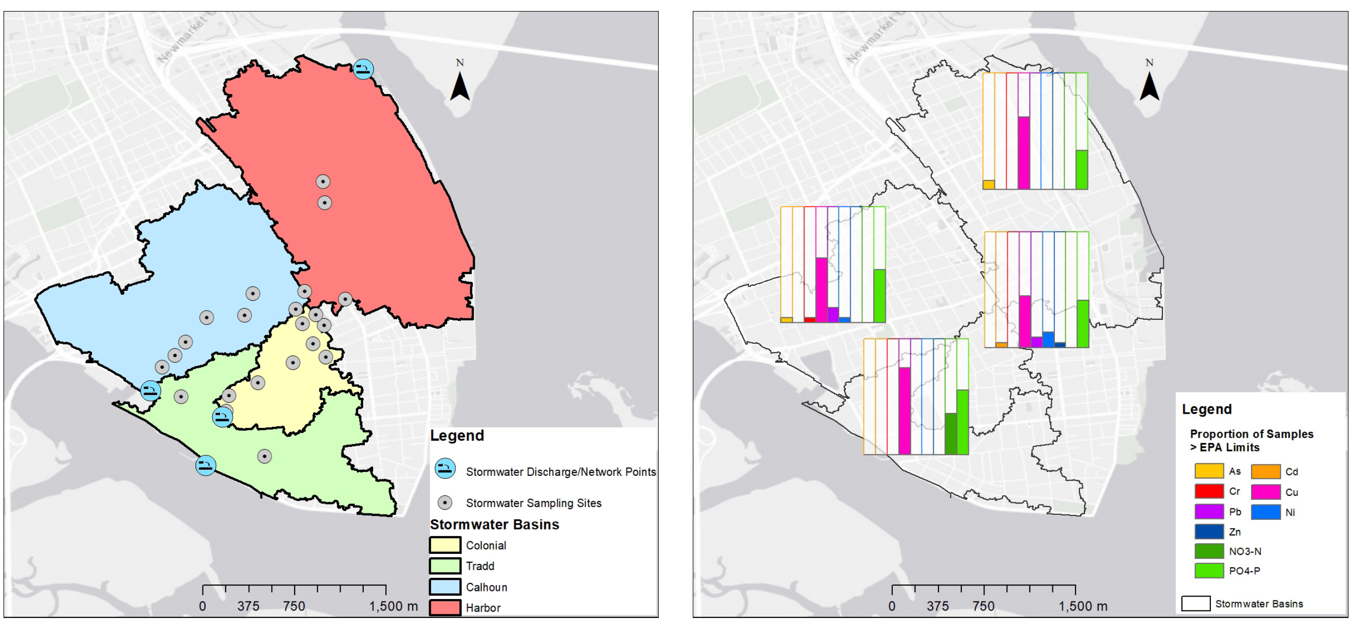

Figure 2. On the left, stormwater basins (four subwatersheds) delineated using lidar-derived digital elevation model (DEM) analysis. The city of Charleston's stormwater discharge outlets are also shown. On the right, the major trace metal and nutrient contaminants are highlighted in each of these watersheds. The bars indicate the percentage of samples that exceeded a US EPA standard or recommendation. Enterococci data are not shown here as all samples in all subwatersheds exceeded US EPA standards. Map data sources: USGS, SC DNR, and the city of Charleston.

Table 1. The city of Charleston's interactive zoning map was used in conjunction with digitized basins from their master flood plan to describe the four areas sampled for this study.

Subwatershed	Area	Commercial	Residential	Industrial
	km^2	%	%	%
Colonial	0.6	24.7	75.3	0
Tradd	0.8	6.6	93.4	0
Calhoun	1.5	41.9	56.6	0.5
Harbor	1.4	32.6	32.4	35

Note. Residential land includes all land zoned as single-family, double-family, mixed-use residential, diverse residential, and residential offices.

Table 2. Enterococci statistics for the four subwatersheds.

Subwatershed	*n*	Minimum	Mean	Median	% RSD	% High
	MPN per 100 mL					
Colonial	21	2,500	17,850	24,196	44	52
Tradd	10	860	14,492	17,697	66	30
Calhoun	21	1,530	17,637	24,000	47	48
Harbor	11	8,010	22,432	24,196	22	82

Note. n is the number of samples collected, *Minimum* refers to the minimum MPN value determined over the entire sampling period, and *Mean* and *Median* refer to statistics conducted on the dataset over the entire sampling period. % High refers to the proportion of samples exceeding maximum high detection limit of 24,196 MPN per 100 mL. Every sample collected exceeded the SCDHEC's recreational standard for enterococci in marine waters of 104 CFU per 100 mL for a single sample.

named for the streets and landmarks in their area; the Calhoun subwatershed flows into the Charleston Marina, the Tradd subwatershed into the mouth of the Ashley, the Colonial subwatershed to Colonial Lake, and the Harbor subwatershed to the Cooper River. The land use in each basin was determined from the city of Charleston's zoning maps (https://gis.charleston-sc.gov/interactive/zoning/) and was categorized as residential, industrial, or commercial. Table 1 shows the percent of each subwatershed zoned for these uses. All sites were considered "urban" or "built-up," and the most common land uses within this urban environment are residential and commercial. Only the Harbor Basin had a significant proportion of industrial land, as a result of the Charleston Ports Authority cargo terminal along the Cooper River. The area of each subwatershed is listed in Table 1, and the subwatersheds averaged 1.1 km^2 in size. Significant pooling of stormwater runoff was observed at areas of low elevation in all subwatersheds.

STORMWATER QUALITY

The enterococci levels in every water sample collected were higher than any state or federal recreational water quality standard (Table 2). The average (arithmetic mean) of enterococci concentrations across all stormwater samples was 18,046 MPN per 100 mL. Even with 100-fold dilution, many water samples from many sites frequently exceeded the upper range on the Enterolert test method (i.e., every well in the Quantitray fluoresced under UV light). In the Harbor subwatershed, 82% of samples had at least 24,196 MPN per 100 mL. The average concentration in the Harbor subwatershed (22,432 MPN per 100 mL) was higher than the rest of the subwatersheds (Figure 3). The Tradd subwatershed has both the lowest average concentration (14,492 MPN per 100 mL) and the lowest percent of samples exceeding the detection limit (30%). However, there was large variability in enterococci concentrations, with some samples having as few as 860 MPN per 100 mL, and as such there was so much overlap between groups that no statistically significant differences between subwatersheds could be determined.

Every sample collected for this study had enterococci concentration higher than the SCDHEC recreational standard (S.C. Code Sections 48-1-10 et seq.) of 104 CFU per 100 mL (note that CFU and MPN values are equivalent). The average MPN counts were comparable to coastal stormwater studies in North Carolina, suggesting that high concentrations of fecal indicator bacteria are likely prevalent in the southeastern coastal plain (Parker et al. 2010).

We analyzed the "first flush" effect, where measured concentrations of an aqueous contaminant increase during initial stages of a storm following a dry period (Hathaway and Hunt 2011). This was not observed for enterococci concentrations in stormwater runoff; in fact, the opposite was true. A 2-sample T-test showed that the mean enterococci concentration of samples collected after 3-day dry periods was significantly lower than in those collected after more than 0.5 cm antecedent rainfall in the 3 days preceding (p-value = 0.013). Rain volume during the event itself (during the 24-hour period, which included sampling) did not appear to be related to the concentration of enterococci in stormwater runoff, unlike antecedent rainfall. Figure 3 highlights data collected with and without antecedent, and it appears that rainy days preceding sampling correlated positively with higher enterococci concentration. Average enterococci concentration was also observed to be higher in the late summer and fall. The average enterococci concentration for all our sites in September 2016 was 24,196 MPN per 100 mL and dropped to below 15,000 MPN per 100 mL from January 2017 until May 2017. By July 2017, the average enterococci concentration for all sites was comparable to the early fall 2016 high concentrations, before dropping again by January 2018. It was determined that the highest enterococci concentration in runoff was present after antecedent rainfall and during the summer and fall. Excessive enterococci concentrations were geographically distributed so that all subwatersheds exceeded US EPA regulations on enterococci concentrations for recreational water, although the Tradd Basin had lower concentrations of enterococci than other subwatersheds.

Trace metals were detected in all stormwater samples and at most sites. Of all trace metals that were analyzed, we consistently detected As, Cd, Cr, Ni, Pb, V, and Zn in most samples. Summary statistics for the detected concentrations of trace metals in stormwater sites are presented in Table 3. The relative standard deviation (RSD) of trace metal concentration within these samples was very high, indicating high variability. Table 3 also lists the maximum detected concentration of these trace metals and compares these concentrations to the US EPA's chronic saltwater toxicity limits (US EPA 2020). Maximum detected trace metal concentration exceeded the toxicity limit of all trace metals, except V. For example, the average concentration of Cu among all samples was 24.0 μg L^{-1}, which exceeds the US EPA's chronic saltwater toxicity index of 3.1 μg L^{-1}, and therefore, high Cu levels in Charleston's stormwater runoff would be a concern to aquatic life in the Charleston Harbor. Copper in the stormwater samples exceeded the chronic saltwater toxicity index for >45% of all samples collected in all subwatersheds. Five out of the remaining six trace metals exceeded the toxicity index in the Calhoun and Colonial subwatersheds. The Harbor and Tradd subwatersheds had either one or no trace metals (other than Cu) that exceeded the toxicity index. The spatial distribution of samples exceeding toxicity standards is plotted in Figure 2.

Previous studies positively correlate trace metal contamination in stormwater runoff to automobile traffic in the watershed (Aryal et al. 2010; Ma et al. 2016); hence, traffic data was also considered alongside trace metal data in stormwater. Annual daily traffic volume (AADT volume) data for the Charleston peninsula (SCDOT 2020) was used for the quantitative evaluation of the relationship between traffic and trace metal concentrations. Additional factors used were 3-day antecedent and event (24-hr) rain volume. Principal component analysis (PCA) was performed on the trace metal, precipitation, and traffic data to determine potential trends. In Figure 4, the first two principal components (PC1 and PC2), which accounted for less than half of the variance, and the correlation vectors for all variables studied are shown. Vectors within each quadrant are strongly correlated, indicating that traffic volume, 24-hr rain, and the trace metals As, Cd, Ni, and Pb are all positively correlated. Since vectors in adjacent quadrants are weakly correlated, there is a weaker but positive correlation between 24-hr rain and the other trace metals. Likewise, the data appears to support that 3-day antecedent rainfall is weakly, but positively, correlated with some trace metal concentrations (As, Cd, Pb, and Ni) and negatively correlated with the other trace metals (Cu, V, Zn, and Cr); that is, rain in the days preceding sampling is related to lower concentrations of these trace metals in runoff: a first flush effect. Also note that the trace elements that appear in each quadrant (e.g., As, Cd, Pb, and Ni) are likely to appear in water samples together and to a lesser degree with Zn, Cu, V, and/or Cr. Land use (industrial vs. residential vs. commercial) was not observed to significantly affect trace metal concentrations and was not included in the PCA biplot, but as illustrated in Figure 2, the Colonial and Calhoun basins were most likely to have samples exceeding toxicity standards for Pb and Ni. In these basins, 8 and 10 samples, respectively, were taken from sites with more than 5,000 average daily vehicles, while the Harbor and Tradd basins contained only one such sample each. The enterococcus data was also not included in the PCA analyses as every sample tested at every site had concentrations that significantly exceeded the SCDHEC's recreational standard.

NO_3^- and PO_4^{3-} concentrations were used as nutrient chemical proxies in the stormwater samples and were averaged across each subwatershed. NO_3^- was present in >60% of the samples in all subwatersheds, while PO_4^{3-} was present in >35%

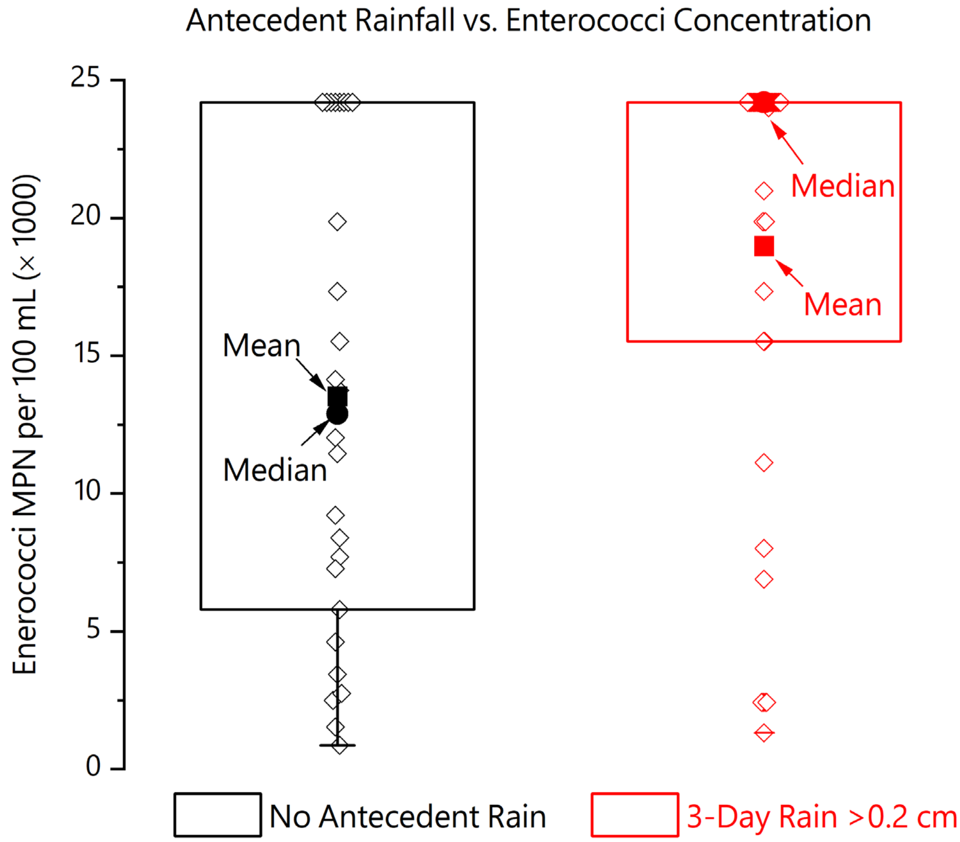

Figure 3. Box plot of enterococci concentrations in the stormwater runoff samples without (left) and with (right) antecedent rain (3 days prior to sampling). Mean (solid squares) and median (solid circles) values are also shown for each set of data. Overall, antecedent rainfall is positively correlated to the concentration of enterococci in stormwater runoff.

Table 3. Major trace metals of interest that were detected in the stormwater samples. Their concentrations varied significantly as shown in % RSD values. Not all trace metals were detected in every sample, as indicated below, and only concentrations that exceeded 0.1 μg L^{-1} were detected and reported. Detected concentrations were compared to the US EPA's chronic saltwater (SW) toxicity standards. Concentrations that exceeded the chronic saltwater toxicity are highlighted in red.

	As	Cd	Cr	Cu	Ni	Pb	V	Zn
Maximum, μg L^{-1}	71.6	16.3	82.1	146.2	26.2	41.5	14.0	142.4
Mean, μg L^{-1}	4.8	0.7	7.4	24.0	2.5	4.8	6.0	31.1
Median, μg L^{-1}	1.3	0.2	3.3	12.1	1.1	2.4	5.0	22.5
% RSD	254	357	174	124	180	156	70	92
% detection	75	60	76	69	79	84	43	84
SW Tox Std, μg L^{-1}	36	7.9	50	3.1	8.2	8.1	–	81

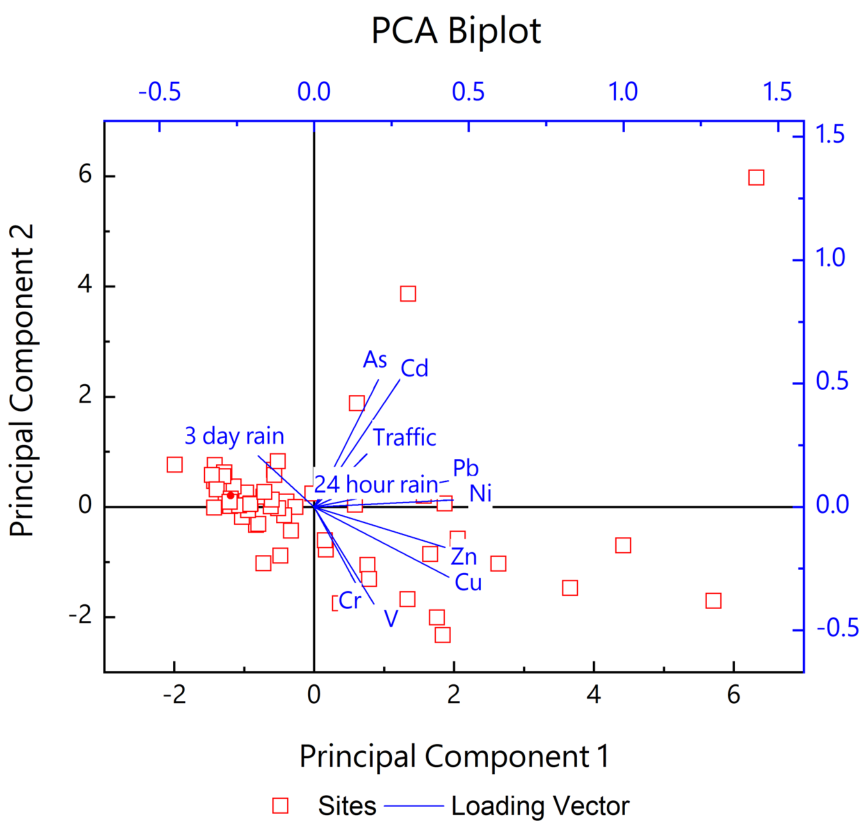

Figure 4. Principal components biplot showing sample clusters and loadings (vectors) between principal components 1 and 2. The data included for these analyses include trace metal concentrations, event rainfall, antecedent rainfall, and traffic counts. Vectors within each quadrant are strongly correlated, while vectors in the diametrically opposite vectors are inversely correlated. Vectors in adjacent quadrants are weakly correlated.

Table 4. Summary statistics of NO_3^- –N and PO_4^{3-} – P concentrations in stormwater samples from the four subwatersheds.

		NO_3^-–N, mg L^{-1}				
Subwatershed	***n***	**Maximum**	**Mean**	**Median**	**% RSD**	**% Detection**
Colonial	22	0.55	0.23	0.27	81	67
Tradd	9	3.5	0.94	0.35	148	78
Calhoun	23	0.50	0.28	0.32	53	83
Harbor	12	0.50	0.28	0.29	57	82
		PO_4^{3-}–P, mg L^{-1}				
Subwatershed	***n***	**Maximum**	**Mean**	**Median**	**% RSD**	**% Detection**
Colonial	22	1.21	0.25	0.00	165	38
Tradd	9	3.46	0.71	0.36	148	55
Calhoun	23	1.47	0.23	0.00	118	48
Harbor	12	0.50	0.15	0.00	139	36

Note. NO_3^- and PO_4^{3-} concentrations were converted to $NO_3^- - N$ and $PO_4^{3-} - P$ concentrations to allow comparisons to US EPA standards. All concentrations are in mg L^{-1}, *n* is the number of samples analyzed, % RSD is relative standard deviation in all samples measured within the subwatershed, and % Detection refers to the percentage of samples that contained detectable concentrations. Detected concentrations were compared to the US EPA's nutrient criteria. Concentrations that exceeded the nutrient criteria are highlighted in red.

of all the samples analyzed (Table 4). The concentrations ranged from 0.91-15.7 mg L^{-1}, while concentrations ranged from 0.94-10.6 mg L^{-1}. The Tradd subwatershed had the highest average concentration for both anions, but it also had higher variability (% RSD) between sample concentrations. The Tradd subwatershed is primarily zoned for residential use (Table 1) and has a higher density of historical homes with immaculately landscaped lawn and garden spaces compared with the other subwatersheds. A t-test did not show a significant change in mean NO_3^- and PO_4^{3-}concentrations after antecedent dry conditions versus 3-day rainfall >0.2 cm.

The US EPA's nutrient criteria recommendations for maximum total N and P in the Southeastern Coastal Plain are 0.9 mg L^{-1} and 0.04 mg L^{-1}, respectively (US EPA 2000). The $NO_3^- - N$ concentration in the Tradd subwatershed was higher than the US nutrient criteria recommendation; however, none of the other subwatersheds exceeded this recommendation on any samples. The mean $PO_4^{3-} - P$ concentration in all subwatersheds was higher than the US EPA's nutrient criteria recommendation.

DISCUSSION

The goal of this study was to evaluate the usefulness of stormwater monitoring in identifying geographically high-risk areas for stormwater runoff pollution. As hypothesized, the urban footprint of the area resulted in significant pollution of the stormwater runoff.

The automatic GIS-based watershed delineation, which relies on high-quality elevation data (DEM), may have inherent artifacts or biases. At the time of this study, only a 2.5-m resolution lidar data was available, and the more recent 1-m resolution lidar data may likely provide additional insights during the watershed delineation (Gillin et al. 2015; Thomas et al. 2017). However, considering the rapid changes that have occurred to the built landscape of peninsular Charleston in recent years, the lidar data would have to be reassessed periodically for changes to the landscapes. Other inherent artifacts and inaccuracies are also reported in the use of various GIS-based watershed delineation methods such as the ArcHydro tool, the Hydrology toolset, and the ArcSWAT tool (Ray 2018). Other researchers may consider a systematic review of the different delineation methods for highly urbanized areas such as Charleston.

Fecal indicator bacteria or FIB (enterococci) levels were very high in stormwater runoff in all subwatersheds, regardless of the predominant zoning within the subwatershed. The most significant cause for impairment of all coastal waters in South Carolina and other similar locations is fecal bacteria (Chen and Chang 2014; Hathaway et al. 2010; SCDHEC 2018). Potential culprits for these high levels are pet waste, wildlife, and failing septic or sewage infrastructure (Steele et al. 2018), though septic infrastructure has been replaced with municipal sewer systems in the Charleston peninsula. The presence of these bacteria poses a significant health risk to residents of these communities who may be exposed to the potentially harmful, pathogen-rich stormwater (Gaffield et al. 2003). Studies have pointed to not only the impairment of the final receiving water bodies (e.g., Charleston Harbor), but also the increased presence of antibiotic-resistant bacteria leading to serious health outcomes (Ahmed et al. 2018; Gaffield et al. 2003; Lee et al. 2020; Scott et al. 2016; Webster et al. 2004). Recent studies also suggest that the risk of human exposure to virulent pathogens such as *Vibrio* is increasing due to climate change–related impacts in coastal regions (Deeb et al. 2018).

Antecedent rainfall had a positive correlation to the presence of enterococci in stormwater and was also observed in other studies (Chen and Chang 2014; Hathaway et al. 2010; Mccarthy et al. 2012; Siewicki et al. 2007). Higher average enterococci concentrations were also observed in late summer and fall, during which time this region generally experiences higher rainfall (Prat and Nelson 2014). Total suspended solids or TSS (not analyzed in this study) are positively correlated with FIB levels, and higher precipitation and strong flowrates generate higher TSS in runoff (Mccarthy et al. 2012; Surbeck et al. 2006). Some studies have shown that the "first-flush" effect may not generate high concentrations of FIB in stormwater (Hathaway and Hunt 2011). It was suggested in these studies that antecedent climate conditions, including atmospheric moisture conditions, positively correlated with the survival rates of bacteria. Larger bacteria peaks are often associated with runoff associated with storms that have antecedent rainfall.

The trace metals observed in the stormwater runoff are some of the commonly observed nonpoint source pollutants in urban runoff, and the trends observed in this study align with reported data in other studies (Baalousha et al. 2019). In this study, the average trace metal concentrations did not appear to be excessive based on the US EPA's recommended ecological standards; however, these lower concentrations may be misleading. We analyzed trace metals in filtered water samples (< 0.22 μ) and not in the composited stormwater samples, which would account for trace metals associated with TSS and other particles such as organic matter. Trace metals strongly bond with a variety of environmental surfaces, including clay minerals, mineral oxides, and organic surfaces (Djukić et al. 2016; Herngren et al. 2005; Vulava et al. 2019). These trace metal–contaminated solids can remain suspended in the final receiving bodies, depending on the specific gravity of the suspended solid, and eventually settle out of the water column into the bed sediment. It is highly plausible that the overall chemical contaminant loads in the stormwater runoff is significantly higher than the concentrations reported in this study. In future studies, it would be useful to measure trace metal concentrations in bulk stormwater samples. The presence of trace metal–contaminated estuarine sediment in the Charleston estuary is well documented and was reported to be higher near urban watersheds (Sanger et al. 1999). In addition, these trace metals may potentially enhance antibiotic resistance in bacteria, including enterococcus and *Vibrio* bacteria. Baker-Austin et al. (2006) found that the presence of trace metal contamination is a chronic and recalcitrant selection pressure with both environmental and clinical importance that may contribute to the maintenance and spread of antibiotic resistance in aquatic environments.

Nutrient pollution has long been identified as a significant degrader of coastal water systems across the US and the world, resulting in eutrophication, harmful algal blooms, shellfish poisoning, and fish kills (Howarth et al. 2000). Typical sources in urban watersheds include lawn fertilizer use and subsequent runoff of excess or improperly applied fertilizer (Toor et al. 2017). Recent studies demonstrate that nearly 80% of P and 20% of N from lawn fertilizer application are part of stormwater runoff in urban watersheds (Hobbie et al. 2017). Higher nutrient inputs were observed in the highly residential Tradd subwatershed; however, higher P concentrations were observed in all subwatersheds. Nutrient ions can also be associated with higher TSS in surface runoff due to the charged nature of the nutrient ions the environmental particles (Sparks 2003; Vaze and Chiew 2004; Wijesiri et al. 2019). Regionally, high concentrations of contaminants associated with stormwater runoff also deposit a wide range of contaminants into the ubiquitous stormwater retention ponds in the region (Beckingham et al. 2019; Cotti-Rausch et al. 2019).

Currently, the main strategy of managing stormwater in the general study area is to quickly pump the water into Charleston Harbor, which has reduced severe flooding in the area. However, flooding still occurs periodically following short and intense storms, especially during spring tides, and can overwhelm the area (Musser et al. 2016). Coastal regions also experience sunny day or "nuisance" flooding due to higher-than-normal spring tides (typically MLLW >7 ft) or King Tides (Román-Rivera and Ellis 2018) and increasingly higher seawater thermal expansion (Widlansky et al. 2020). In the last several years, such flooding has increased significantly in the Charleston peninsula and in other similar coastal areas (Morris and Renken 2020). Predicted and observed tidal data obtained from https://mycoast.org/sc show that King Tides are increasing in frequency near the Charleston peninsula, with more than 70 observations of MLLW >7 ft each year from 2016 to 2018. The resulting higher coastal water table elevations can potentially lead to increased backup of stormwater during coincident precipitation events.

The flooding-related problems also predominantly affect lower-income and minority communities in Charleston, as is the case in other urban areas of the US (Montgomery and Chakraborty 2015). More effective best management practices (BMPs) and strategies need to be incorporated into sustainable and socially equitable stormwater management plans (Ahmed et al. 2019; Allen et al. 2019; Prudencio and Null 2018). The data collection and mapping framework used in this study can be used in the development of effective plans.

CONCLUSIONS

There is widespread contamination of stormwater runoff in urban areas such as the city of Charleston. Fecal bacteria are present at extreme levels and can pose a significant health risk to local communities. Trace metals and nutrient contamination are also present in the stormwater runoff at relatively high concentrations and can potentially enhance the antibiotic resistance of the fecal bacteria. Collectively, these contaminants, as well as other persistent and emerging contaminants that were not monitored in this study (e.g., persistent organic contaminants, microplastics), pose a significant threat to the coastal ecosystems. The resulting economic impact could be detrimental to important ecosystem services, such as recreation and seafood safety within the region. Stormwater runoff will add to the increasing coastal flooding, which is expected to only become worse due to the rapidly changing climate; therefore, innovative and sustainable solutions have to be investigated. Traditional strategies to reduce flooding and managing stormwater require significant infrastructure improvements and overcome significant technical challenges. However, protecting public health by reducing exposure to stormwater runoff and associated nonpoint source pollution is paramount.

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