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Chemical characterization of urban stormwater: Traditional and emerging contaminants

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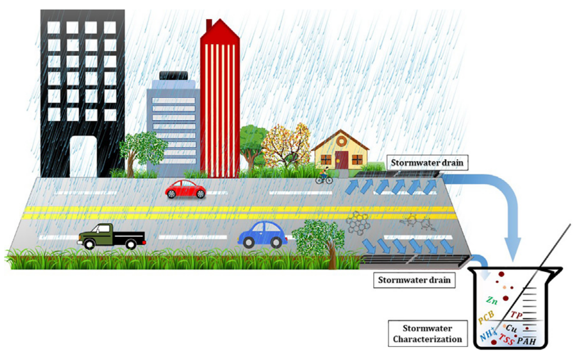
HIGHLIGHTS

- Extensive characterization of urban stormwater quality including traditional and emerging pollutants
- Database and literature review analysis include thousands of data points.
- Particle associated and dissolved contaminants are included.
- Research gaps include trace pollutants, spatial variability, and particle chemistry.

GRAPHICAL ABSTRACT



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ABSTRACT

Increases in urbanization have led to increased stormwater runoff and mobilization of pollutants from urban watersheds. Discharge of these pollutants often leads to contamination of receiving water bodies. Chemical characterization of urban stormwater is necessary to gain deeper insights into the ecological impacts of urban runoff and to evaluate parameters that influence possible treatment technologies. This study assessed stormwater event mean concentrations and particle size fractions from field studies reported in national/international stormwater quality databases (The National Stormwater Quality and The Best Management Practices databases) and peer-reviewed literature. This characterization of urban stormwater includes statistical evaluation of probability distribution, consideration of dissolved and particulate-bound pollutants and focuses on partitioning and speciation behavior. Solids, nutrients, metals, organic pollutants, and bacterial pathogen indicators were evaluated. A significant fraction of stormwater phosphorus, metals and organic pollutants are particle-bound. Results from the speciation of metals demonstrated that metals are predominantly present as either inner-sphere or electrostatic complexes with dissolved organic matter. This study provides a comprehensive overview of the myriad pollutants found in urban stormwater and provides a starting point for addressing ubiquitous and emerging contaminants. Finally, research needs for further detailed stormwater characterization were identified.

1. Introduction

1.1. Overview

Rapid increase in global urbanization is producing multiple environmental challenges, one being the impacts of urban stormwater runoff (US EPA, 2015a). In a setting with natural land cover, rainwater will primarily infiltrate through the soil surface or return to the atmosphere via evapotranspiration. During the urbanization process, the fraction of impervious land area, including driveways, parking lots, roadways, and rooftops, increases resulting in increased volumes and velocities of surface runoff (Goonetilleke et al., 2005; Hsieh et al., 2007). Rainwater is not contaminant free as it contacts impervious surfaces and becomes stormwater, but further mobilizes a broad range of pollutants including particulate matter, nutrients, oil and grease, toxic organic compounds, metals, and microorganisms.

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This runoff discharges into local receiving water bodies, frequently causing water quality impairment and posing harm to aquatic (and possibly human) life. Stormwater pollutants originate from natural and anthropogenic activities in the urban watershed thus making stormwater a large contributor to non-point source pollution (US EPA, 2015a).

1.2. Stormwater pollutants

While the contaminants present in stormwater vary by region and the degree of urbanization, several contaminants are considered important analytes of special concern because they may be toxic or otherwise negatively impact the health of the receiving water body. Nutrients such as nitrogen (N) and phosphorus (P), are washed from terrestrial surfaces during rain events and can accumulate downstream, possibly leading to eutrophication (US EPA, 2016). Trace metals, such as Cd, Cu, Pb, and Zn, are ubiquitous in urban watersheds, originating from vehicle brake dust, tire wear, or building materials like copper roofing and galvanized fencing (Eriksson et al., 2007; Sakson et al., 2018). While metals like Cu and Zn do not pose a lethal risk to humans at low levels found in stormwater, they can be toxic for many aquatic organisms at trace levels. Organic pollutants are a large and highly diverse category of pollutants. In addition to natural carbon released from the decomposition of living tissue and animal wastes, this category includes anthropogenic compounds such as pesticides, hydrocarbons (i.e., oils, gasoline, etc.), and a variety of industrial, household, and pharmaceutical chemicals (Eriksson et al., 2007; Masoner et al., 2019). Due to their chemical complexity and the sheer number of anthropogenic organic compounds in use, it is generally impractical to test for specific chemicals other than through a research endeavor.

1.3. Stormwater management

Stormwater Control Measures (SCMs) or Best Management Practices (BMPs) are designed and implemented to mitigate the quality and quantity of stormwater to prevent flooding and excess stream flow/velocities and improve overall water quality. SCMs include rainwater harvesting systems, green roofs, bioretention facilities, vegetated swales, permeable surfaces, infiltration trenches, stormwater wetlands, and various pond systems (Davis, 2007; Hatt et al., 2007; Natarajan and Davis, 2015; Wu and Allan, 2018). The functional performance of a SCM will depend on the physical, chemical, and biological characteristics of incoming stormwater such as pH, salts/ionic strength, particulate matter (PM), particle size distribution (PSD), and bacteria (including pathogens) in determining pollutant fate, toxicity, and treatability (Dean et al., 2005). For example, a large portion of phosphorus, trace metals (Cu, Zn, Pb) and organic pollutants are transported primarily via stormwater PM, in turn impacting pollutant partitioning and speciation (Li et al., 2005; Sansalone and Buchberger, 1997b; David et al., 2011; Sansalone and Buchberger, 1997b). Therefore, developing effective SCMs will depend on the aquatic and surface chemistry of stormwater and different particle sizes and PSDs. These characteristics will impact SCM unit operations designed for improving stormwater quality.

1.4. Objectives

Stormwater quality characterization is thus a multivariate analysis that requires large amounts of data from across the world. In this pursuit, several studies have attempted to characterize stormwater and relate pollutant concentrations and/or loads to factors such as land use, daily traffic count, antecedent dry period, and/or fluctuations in weather and climate (Barrett et al., 1998; Lee and Bang, 2000; Thomson et al., 1994; Wu and Allan, 2018). While these studies have made advances in understanding these complex relationships, data gaps remain due to the wide variability in spatial and temporal stormwater/land interactions. Nonetheless, understanding stormwater quality is important to quantifying the environmental and ecological impacts, and treatability, of stormwater.

Given the scale of the issues associated with stormwater management, a central public database of stormwater characterization studies is a valuable tool for public policy development. Such databases exist in the form of the *International Stormwater Best Management Practices database* and the *National Stormwater Quality Database* (https://bmpdatabase.org/).

The overall goal of this study is to utilize these databases and assess collected field data to characterize and summarize the complexity of urban stormwater. These data can provide insights for regulators and practitioners in determining water quality standards and treatment goals, as well as identify data gaps for researchers. A thorough characterization of stormwater is necessary for use in studies of SCM performance that may require the use of a synthetic stormwater. The objectives for this study are: 1) utilizing large datasets and statistical evaluation, summarize constituent concentrations for solids, urban chemical pollutants (inorganic and organic), and pathogens, 2) evaluate both contaminants of concern and background constituents (e.g., pH, alkalinity, natural organic matter) affecting speciation and partitioning of stormwater pollutants, and 3) identifying research needs in stormwater monitoring and characterization.

2. Methodology

2.1. Data collection

2.1.1. Water quality databases

The International Stormwater Best Management Practices database and the National Stormwater Quality Database were used as the primary data sources (https://bmpdatabase.org/). The International Stormwater Best Management Practices (BMP) database has data compiled from >700 stormwater treatment studies and has been operational since 1996; the updated version of the database collected data until 2020. Many SCM field study results have been entered into the database after meeting the requirements for submittal for better reliability of the data. The BMP database provides data entries from the United States, Canada, New Zealand, and Australia where stormwater/SCM research is extensively taking place. The database consists of information for different SCMs, including grass filters, bioretention cells, vegetative swales, infiltration basins and others, across various land uses and climates. In addition, cost, performance efficiency, physical site characteristics, EPA rain zones, and influent and effluent concentrations of constituents are catalogued. Evaluating SCM treatment performance is beyond the scope of our study; therefore, the inflow data were solely pulled for stormwater quality assessment. Detection limits, sample types, sample dates, and inflow event mean concentrations were used. Greater than 98% of the data are from the United States; thus, extrapolation to other global areas where stormwater characteristics may be different should be done with caution.

The National Stormwater Quality Database (NSQD) was developed for stormwater quality characterization in the U.S. by Dr. Robert Pitt in 2001 and primarily contains stormwater data from NPDES (National Pollutant Discharge Elimination System) and MS4 (municipal separate storm sewer system) stormwater permit holders. NSQD database version 4.02 (Pitt et al., 2018), which is now available on the BMP database website, is up to date and presents more data than its older version, with the new version holding data from 200 municipalities and approximately 8744 storm events across the U.S. Data entries pertaining to the influent characteristics of stormwater across various land covers are included, unlike the BMP database where information related to the performance aspect of the SCMs is the central focus.

2.1.2. Literature

This study also examined dissolved pollutants data derived from a multitude of stormwater field research papers from peer-reviewed journals for comparison with the data from databases, but were not included in the statistical analysis. Literature was also appraised to include and assess underreported or complex water quality parameters such as particle size distribution of particulate matter and emerging organic contaminants that have little/no/ data in the databases. Literature search included keywords such as urban stormwater quality, highway characteristics, event mean concentrations (EMCs), nutrients, trace metals, organic pollutants, and particle size distribution in stormwater. Data reported before the year 1995 were omitted to focus on more reliable and recent values as analytical methods improved and material bans have been implemented (e.g., lead in paint and gasoline).

The literature was mined for studies that conducted particle size distribution analysis for stormwater sediments. Studies that conducted analysis on settled particles in water bodies or any application other than stormwater were not included. Identified data for this study evaluated particles in stormwater and collection of road-deposited sediments (German and Svensson, 2002; Lau and Stenstrom, 2005; Sansalone and Tribouillard, 1999).

2.2. Data processing and analysis

The BMP and the NSQ database results were combined before statistical analysis. Some duplicate data were found. The NSQD includes a column in the database that identifies the original source of data, which enabled the exclusion of double counting. Also, data from the BMP database that had a few pretreatment (SCMs in series) steps were excluded from the analysis. Only composite sample data were taken from the databases, as these data represent an accurate estimate of the event mean concentration (EMC - volume weighted constituent concentrations) of the entire storm event. Industrial, rangeland, forested, open space and agricultural land uses, manual (grab) sampling, and data from before 1995 were excluded from the analysis to focus on current urban runoff characterization, and to minimize issues associated with product bans and decreasing methods detection limits.

Prior to estimating the descriptive statistics on the EMCs, percentage of non-detects ("less than" or below instrument detection limits, %BDL) for each parameter was calculated to determine the most suitable method to handle non-detects within these datasets. The EMCs identified as non-detects in the BMP database were listed in the database at half the detection limit. These values were therefore multiplied by 2 to obtain the corresponding original detection limits. The NSQ database reported non-detects without any substitutions, hence the correction was not necessary.

In the combined databases, non-detects for many pollutants, particularly metals and organics, occurred at >15% of the total observations. This many below-detects required a more robust approach for estimating these values over simple substitution methods (USEPA, 2000). Our analysis was limited to water quality parameters with less than 50% non-detects. Since the database data were gathered from various monitoring sites in the US, each dataset contained multiple detection limits. The regression on order statistics (robust ROS) method, as recommended by Bolks et al. (2014) (discussed by Helsel and Cohn (1988) as the "fill-in" probability plotting procedure), was used for datasets with <50% non-detects and with at least three different detection limits. The *Nondetects and Data Analysis* (NADA) for R package developed by Dennis R Helsel (Helsel, 2011) was installed to calculate the plotting positions and the descriptive statistics, and to create the probability exceedance plots for the selected water quality parameters.

RStudio version 1.2.1335 was employed to process and run statistical analyses on the large datasets for all constituents derived from the databases. Probability plots (histograms) for all the constituents were developed for an overall view of the raw data, and to assess normality (central limit theorem). Most data do not follow a normal distribution and were prominently right skewed due to the presence of non-detects, which explains the notable differences between the mean and median values (mean > > median). Therefore, the data was $\log_{10}$ transformed to check for a log-normal distribution. The $\log_{10}$ transformed distributions of some of the analytes do not strictly represent normality either, particularly the values at lower concentrations. This occurred because of the experimental approximations made for EMCs below the detection limits or instrument errors. Pitt et al. (2018) observed similar trends where the data for constituents were not normally distributed even after log transformations. However, datasets with most quantifiable observations followed a log-normal distribution with no special peaks at the tails of the curve, while analytes with many non-detects showed a skewed distribution even after a log transforming the data. Violin plots for water quality parameters with a large number of data points and few non-detects, total nitrogen (TN), total phosphorus (TP), total suspended solids (TSS), and total zinc (Zn T) were constructed to assess log-normal distributions of the data (Fig. 1). Following these trends, the geometric mean (or geomean), geometric standard deviation (GSD) and geometric standard error (GSE) were calculated for each of the constituents, along with other summary statistics, as measures that represent typical stormwater concentrations.

2.3. Metals speciation

Visual MINTEQ (version 3.1), a geochemical equilibrium software, was used for modelling of metal speciation in stormwater. The NSQ and the BMP databases did not provide information about the type of PM solid phases. Therefore, only the aqueous (dissolved) metal concentrations were considered for the speciation analysis. By including dissolved metal concentrations as opposed to total metal concentrations, a clear distinction was predefined which account for partitioning attributes related to speciation. Important input water quality parameters from the databases included dissolved metal concentrations, pH, temperature, and conductivity. MINTEQ requires input on Ionic Strength (I) which was estimated from electrical conductivity (κ) using (Snoeyink and Jenkins, 1990):

$$I\ (\mathrm{mol/L}) = 1.6*10^{-5}*\kappa\ (\mu\mathrm{S/cm}) \quad (1)$$

Information on inorganic ligands such as chlorides (Cl^-), nitrates (NO_3^-), phosphates (PO_4(-III)), and carbonates (CO_3(-II)), and organic ligands, measured as total or dissolved organic carbon, were available in the databases.

Dissolved organic matter (DOM) is heterogenous and exhibits a high affinity for metals. Due to its heterogeneity and presence of multiple functional groups, DOM does not exhibit unique complexation constants. The advanced non-ideal competitive adsorption (NICA)-Donnan model was chosen from MINTEQ to estimate the complexation with DOM. This model is an isotherm model, which allows for simulating metal ions complexation with humic substances, taking into account the heterogeneity of DOM and the inherent electrostatic interactions of ions with the DOM sites (Benedetti et al., 1995; Kinniburgh et al., 1999). Recent studies in China have attempted to characterize the fundamental structures of DOM specifically in stormwater discharging from urban regions (Zhao et al., 2015; Chen et al., 2017; Liao et al., 2021). These studies observed that stormwater DOM is primarily comprised of humic-like substances with high molecular weights originating from local and non-local sources. This being the best information available, DOM was categorized as humic acid in the MINTEQ software for the metals speciation analysis.

The NICA-Donnan model also allows for carboxylic and phenolic site interactions with cations. The MINTEQ software uses the recommended generic model parameters formulated by Milne et al. (2001, 2003) specifically to fit this model. Furthermore, alkalinity was considered as a representative of carbonate species, and orthophosphate (OP) for phosphates.

3. Results: stormwater quality characterization

3.1. Traditional water quality constituents

Traditional water quality parameters were derived from the databases as part of the stormwater characteriztion and for support of other water quality analyses. For example, Oil and Grease (O&G), Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), Total Organic Carbon (TOC), pH, and hardness are important to define stormwater quality and impacts. Other parameters such as alkalinity, Cl^-, and conductivity were needed for metals speciation analysis. Table 1 provides summary statistics for these general water quality parameters from the combined databases.

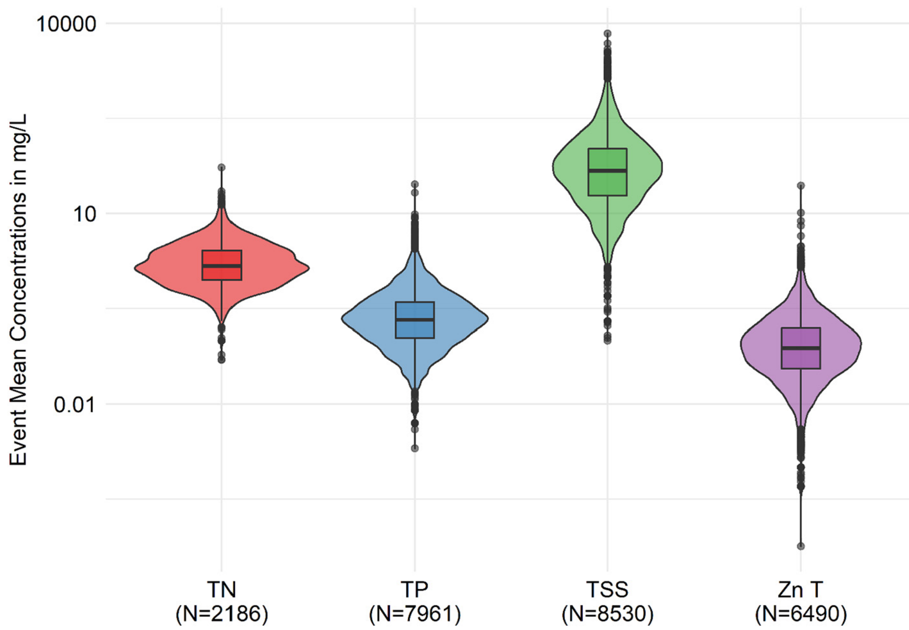

Fig. 1. Violin plots with included boxplots of TN, TP, TSS and Zn T show log-symmetric distributions of the analytes based on the event mean concentrations (EMCs) data from the BMP and NSQ databases.

Geomean and median values were the most similar and represent concentrations that are likely to be present in urban stormwater.

The statistical evaluation showed that stormwater pH values were consistent among studies with mean, median and geomean at 7.1, indicating that the stormwater alkalinity can buffer acidic rainwater (Kuang and Sansalone, 2011). Predictably, the datasets showed huge variability in the range of other constituent EMCs due to stormwater heterogeneity. Background constituents including pH, ionic strength (conductivity), and DOM were tagged as critical factors since they can influence the fate and transport of stormwater pollutants. For example, solution pH and DOM tend to have a greater influence on the trace metal speciation as compared to inorganic ligands (e.g., carbonates, phosphates, chloride) and other water quality parameters (Dean et al., 2005; Gnecco et al., 2008). Understanding the complex chemical matrix in stormwater can provide valuable insights into stormwater pollutant speciation, thereby improving treatment designs tailored to the stormwater characteristics.

3.2. Particulate matter

3.2.1. Solids

PM ranges from colloidal size to gravel aggregates (Sansalone and Kim, 2008). Operationally, particles that can removed by filtration using a 0.45 or 0.2 μm filter are defined as 'suspended solids', which are insoluble and act as mobile substrates (Sansalone and Kim, 2008). Pollutants of size < 0.2 μm are operationally classified as 'dissolved', although they include colloidal material. PM in stormwater can either be measured as total suspended solids (TSS) or suspended sediment concentration (SSC). TSS is a common wastewater measurement, whereas SSC is more applicable to stormwater. The former has been shown in studies to underrepresent the PM concentration in stormwater because of sampling variability, whereas SSC uses a whole-volume sample analysis that reduces the error associated with sampling (Clark and Siu, 2008). Clark and Siu (2008) also observed that SSC was not affected by the particle size distribution of the sample. However, TSS is widely reported as the measure of PM in stormwater studies; this is because detection of SSC practically requires the volume of the entire collected stormwater sample, which limits the analysis of other constituents.

Solids measurements available in the databases include TSS, total dissolved solids (TDS), SSC and total solids (TS). Of all constituents, TSS recorded the second-highest number of observations (>10,000 raw data points, 8530 used in this study). TSS data followed a log-symmetric distribution given the wide range of concentrations being detectable (99.4% of observations were detects, Fig. 1) and the TSS in stormwater was present at relatively high concentrations when compared to other quality parameters. Stormwater concentrations ranged from 0.2 mg/L to 113,860 mg/L for TDS, and 0.1 mg/L to 6900 mg/L for TSS, with geomean values at 76 mg/L and 44 mg/L, respectively (Table 2). As discussed above, the SSC value (geomean) is twice the TSS concentrations.

3.2.2. Particle size distribution

The BMP database contains data for d_{50} (diameter at which 50% of the particles are finer than) for 49 stormwater PM influent samples with only

Table 1
Summary statistics of Stormwater EMCs of General water quality parameters from the International BMP Database and the National Stormwater Quality Database.

Water quality constituents	Units	N	%BDL	Min.	Median	Mean	Max.	Geomean	GSD
Alkalinity as $CaCO_3$	mg/L	772	6.7	0.2	23.3	45.0	753	22.5	3.6
BOD as O_2	mg/L	3409	5.2	0.1	8.0	13.6	433	8.4	2.6
Cl^-	mg/L	1288	15.9	0.2	6.1	37.7	2640	7.3	5.9
COD as O_2	mg/L	2940	3.1	0.5	52.4	79.4	1674	49.7	2.8
Conductivity (κ)	μS/cm	2176	0	0.1	88	210	29,967	97	2.7
DOC	mg/L	1227	0.1	0.5	10	16	113	10.3	2.7
Oil and Grease	mg/L	1326	27.1	0.05	4.0	17.4	2980	3.3	4.5
pH		4219	0	3.5	7.1	7.1	11	7.1	1.1
TOC	mg/L	2005	0.5	0.2	12	18.5	350	12.2	2.5
Turbidity	NTU	1154	0	0.4	19.4	55.6	3680	19.5	3.4

four sites in the same geographical location included. The median value was 40 μm, and the mean was 66 μm. Table 3 shows values given from 10 additional studies that conducted PSDs without any exclusions of particle sizes of interest in stormwater (several studies excluded particle sizes larger than 2 mm). Table 3 was constructed by visually retrieving d_{10}, d_{50}, and d_{90} from graphs provided in the studies; thus, these values are estimations of PSDs in urban watersheds. The majority of the studies were conducted on a road or highway, and the remainder conducted within urban watersheds.

The data of Table 3 are summarized in Table 4. The geometric means of the d_{10} d_{50}, and d_{90}, were 21.0, 242, and 1255 μm, respectively (Table 4). For each, the geometric mean is smaller than the arithmetic mean indicating that the data are skewed to the right. These values show a wider range compared to data from the BMP database. These additional data provide an idea of the variability of stormwater PM PSD (some of which is related to measurement methods employed).

PSD diameter ratios, specifically d_{50}:d_{10}, and d_{90}:d_{50}, were calculated to examine any trend within the PSD for a given storm or catchment area. The mean d_{50}:d_{10} was approximately 10, with a standard deviation of 8; the d_{90}:d_{50} ratio had a mean of 11 and a standard deviation of 12. These standard deviations are considered large. However, the data set is small, and several outliers skew these ratios. The ratios are consistently between 5 and 10 and 0–10 for d_{50}:d_{10} and d_{90}:d_{50}, respectively. Currently, d_{50} is commonly used to compare PSDs, but these ratios may provide a more robust representation of the full PSD.

Variability in stormwater PSDs can be partially attributed to collection and analysis methods. Kim and Sansalone (2008) evaluated PSDs from literature and found that PSDs obtained from road-deposited sediments (e.g., street sweeping, or vacuum collection) exhibited more similar and consistent patterns than PSDs obtained from rainfall-runoff. However, collection of road-deposited sediments is limited in collecting fine PM, where rainfall-runoff is effective at mobilizing the fine PM fractions. Additionally, each analysis method has limitations. Mechanical sieving is consistent for PM >25 μm, while laser diffraction measurements often have upper limits (~2 mm) and may exclude upper portions of the PSD (Kim and Sansalone, 2008).

For stormwater samples, the time between collection and analysis is crucial for accurate particle size distribution. Li et al. (2005) found that stormwater PSD samples need to be analyzed within six hours from collection and should be gently inverted to avoid shearing and sedimentation. Nonetheless, data where the time between sample collection and analysis exceeded 24 h were included as part of our analysis due to the lack of data. For vacuum collected samples, holding time is less important since the sediments are dry thus eliminating potential to aggregate or shear in solution.

PM plays a critical role is many aspects of stormwater quality. It acts as a sink or reservoir for many pollutants, including many toxic compounds, and is responsible for their partitioning attributes. A significant portion of phosphorus, trace metals (Cu, Cr, Pb, Zn) and organic pollutants (PCBs, PAHs) is sediment-bound (Herngren et al., 2005; Zgheib et al., 2011). Hence most SCMs are designed fundamentally to target and capture PM. PM in urban runoff originates from erosion of road surfaces, tire and pavement abrasion, maintenance operations, litter, soil fragments, atmospheric deposition and other anthropogenic activities (Gunawardana et al., 2012; Li et al., 2005).

Table 2
Summary statistics of Stormwater solids EMCs from the International BMP Database and the National Stormwater Quality Database.

Water quality constituents	Units	N	% BDL	Min.	Median	Mean	Max.	Geomean	GSD
SSC	mg/L	694	0.1	1	97	262	7000	99	3.8
TDS	mg/L	3822	2.4	0.2	72	283	113,860	76	3.0
TS	mg/L	205	9.3	12	124	198	2910	121	2.6
TSS	mg/L	8530	0.6	0.1	47	104	6900	44	3.8

Table 3
Overview of PSD from 77 sampling events, expressed as d_{10}, d_{50}, and d_{90}, as well as collection method, and PSD analysis used. The values were compiled visually from PSD graphs from the 10 individual studies shown in the table. All studies were located in urban watersheds.

PM diameters (μm)						
d_{10}	d_{50}	d_{90}	n	Collection Method	PSD Analysis	Reference
125	600	>2000 [b]	2	Vacuum	Mechanical sieving	(German and Svensson, 2002)
9.14	256.3	1303	21	Composite sample	Laser diffraction	(Jartun et al., 2008)
95	400	1800	6	Grab sample	Mechanical sieving	(Vaze and Chiew, 2004)
7	43	400	1	Composite sample	Laser diffraction	(Kim and Sansalone, 2008)
20	233	4000	1			
25	300	10,000	1			
6	29	200	1			
30	247	7000	1			
10	69	2000	1			
15	106	4000	1			
7	58	2000	1			
70	200	1500	7	Vacuum [a]	Mechanical sieving	(Lau and Stenstrom, 2005)
10	100	400	6	Grab sample	Laser diffraction	(Li et al., 2005)
30	150	350	2	Composite sample [a]	Laser diffraction	(Pimiento et al., 2018)
100	600	3000	8	Composite sample	Mechanical sieving	(Sansalone et al., 1998)
100	600	5000	5			
80	500	5000	2	Composite sample	Mechanical sieving	(Sansalone and Buchberger, 1997a)
NR	1000	6000	2			
90	600	3000	5			
65	450	2000	1	Vacuum [a]	Mechanical sieving	(Sansalone and Tribouillard, 1999)
65	600	>5000 [b]	1			
150	1200	>5000 [b]	1			

(a) time from collection to analysis not stated, (b) size estimated to be 20% larger than largest mesh size. NR – not reported.

3.3. Nutrients

3.3.1. Nitrogen

Major and dissolved inorganic nitrogen species in stormwater are present as oxidized N as nitrate (NO_3-N) and nitrite (NO_2-N) and reduced N as ammonium (at relevant stormwater pH = 6–8, ammonia is predominantly present as NH_4^+-N (Stumm and Morgan, 2012)). Total Kjeldahl Nitrogen (TKN) is an operationally defined measurement equal to the total organic nitrogen (TON) plus ammonium (NH_4-N) thus TON is the difference between TKN and ammonium. Also, organic nitrogen can exist both in dissolved (DON) and particulate (PON) forms. Li and Davis (2014) reported PON and DON stormwater concentrations at an overall median of 0.93 mg/L and 0.25 mg/L respectively, indicating a large portion, 79%, of total organic nitrogen to be particulate. Breakdown of nitrogen species:

$$\text{Dissolved Inorganic Nitrogen (DIN)} = [NO_3{-}N] + [NO_2{-}N] + [NH_4{}^+{-}N] \quad (2)$$

$$\text{Total Kjeldahl Nitrogen (TKN)} = [NH_4{}^+{-}N] + TON \quad (3)$$

Table 4
Summary statistics of PSD data gathered from 10 studies in Table 3, including range, mean, standard deviation, and geometric mean of the d_{10}, d_{50}, and d_{90}.

Parameter	Particle sizes (μm)			
	Min	Mean	Max	Geomean
d_{10}	6	40.5 ± 41.2	150	21.0
d_{50}	21	321 ± 224	1200	242
d_{90}	200	1901 ± 1853	10,000	1255

$$\text{Total Organic Nitrogen (TON)} = \text{Dissolved Organic N (DON)} + \text{Particulate Organic N (PON)} \quad (4)$$

$$\text{Total Nitrogen (TN)} = \text{TKN} + [NO_3{-}N] + [NO_2{-}N] \quad (5)$$

The databases contain nitrogen data for TKN, NO_3-N + NO_2-N, NO_3-N, NO_2-N, NH_4^+-N, TON, and TN, with TKN, NO_x and TN as the most commonly reported constituents. The number of individual data entries for all the species were different, indicating that datasets were derived from distinct populations (storm events) and implies that not all studies report nitrogen species together. Some NO_3-N and NH_4^+-N data did not have concentrations reported in terms of N and were corrected before performing any statistical analysis. Limited data for particulate nitrogen (PN) were available, primarily from a single site with commercial land use in Florida and had 19 data points or storm events with a geomean 0.38 mg/L and hence was not included in the data analysis. Non-detects of all the nutrient (N and P) species, irrespective of their %BDL, were treated with the ROS method for estimating non-detects, even though the percentage of BDL was not significantly high for many of these species (Table 5), because each of these datasets carry at least three distinct detection limits. No estimation method was needed for TON. The geomean values for individual analytes (NO_3-N, NO_2-N, NH_4^+-N) and combined (TKN, TON, TN, NO_3-N + NO_2-N) were calculated and compared. The geomean values of individual N species: NO_3-N, NO_2-N, NH_4^+-N are 0.48, 0.05, and 0.25 mg/L respectively. On the other hand, NO_3-N + NO_2-N, TKN, TON, and TN concentrations, respectively, have geomean values of 0.44, 1.32, 0.56, and 1.5 mg/L, indicating that mass balances are not met.

The geomean of TON is 0.56 mg/L (Table 5), which is 48.1% lower than the calculated value of TON using Eq. (3) (1.08 mg/L) and the data for the combined [NO_2-N + NO_3-N] is 17% less than the sum of the individual species. TN at 1.5 mg/L is 18.9% smaller than the geomean data (1.85 mg/L) using Eq. (5) with individual species and 14.8% smaller if combined [NO_2-N + NO_3-N] was used (1.76 mg/L).

The number of observations for TON were much fewer when compared to the other nitrogen species, suggesting it as less reliable when compared to the other species (Table 5); TKN and NH_4-N had the most measurements. Therefore, to arrive at a recommended value for TON, Eq. (3) was used to estimate TON at 1.08 mg/L.

Determining values for TN and NO_x was more challenging, since in both cases the sum of the individual species was greater than the composite measurements. Therefore, the values of the individual species were used for the recommended values. More sampling data that have complete N speciation is needed to provide higher confidence in overall N speciation and mass balances.

With this analysis, the percentage breakdown of the N species from the databases included approximately 58% organic (TON) and 42% dissolved inorganic nitrogen (NH_4^+-N + NO_3-N + NO_2-N) content (Fig. 2). The major species are NO_3-N and TON, and the minor species are NO_2-N and NH_4^+-N. The calculated TON and TN of 1.08 and 1.85 mg/L are shown in Fig. 2. Based on the limited available data, the geomean of DON is 0.92 mg/L from 245 entries, which placed the dissolved organic fraction at approximately 85% of the calculated TON, with 15% being particulate matter.

3.3.2. Phosphorus

As with nitrogen, phosphorus is apportioned into dissolved and particulate fractions. Typical phosphorus constituents include total phosphorus (TP), total dissolved phosphorus (TDP), and soluble reactive phosphorus (SRP), the latter considered as equivalent to orthophosphate (OP), which is the inorganic dissolved form of phosphorus (PO_4(-III)). Of the P species, orthophosphate is of highest concern as it is more readily available for biota to metabolize and comprises most of the soluble phosphorus (Dierberg and DeBusk, 2008; Yang and Toor, 2018). The speciation of OP depends chiefly on the pH of the runoff, where a minor shift in pH can distribute OP forms between $H_2PO_4^-$ and HPO_4^{2-} at relevant environmental conditions (pH = 6 to 8, pK_{a2} = 7.2; (Stumm and Morgan, 2012; LeFevre et al., 2015). Under acidic conditions P sorption to PM decreases, although urban stormwater typically has buffered conditions and sorption effects do not shift noticeably. Particulate phosphorus (PP) and dissolved organic phosphorus (DOP) can be attained by using mass balances and the values of other phosphorus species (Eqs. (6) and (7)). PP fractions can significantly vary depending on site-specific characteristics and studies have found PP to be the predominant form of stormwater phosphorus (Berretta and Sansalone, 2012; Liu and Davis, 2014). Breakdown of phosphorus species:

$$\text{Total Phosphorus (TP)} = \text{TDP} + \text{PP}; \quad (6)$$

$$\text{Total Dissolved Phosphorus (TDP)} = \text{OP} + \text{DOP} \quad (7)$$

A subset of OP data was reported as PO_4 and corrected to units as P before calculating the descriptive statistics. TP, TDP and OP stormwater event mean concentrations ranged from 0.002 to 28.9 mg/L (geomean = 0.2 mg/L), 0.001 to 19.3 mg/L (geomean = 0.09 mg/L), and < 0.001 to 83.2 (geomean = 0.03 mg/L), respectively, in the databases. Data collected from approximately 30 literature studies with mixed land uses note a TP minimum of 0.01 mg/L and a maximum of 1.2 mg/L, which is a tighter range than that obtained from the databases. However, the TP geomean value of 0.2 mg/L from the databases is appropriately within the range of 0.01–1.2 mg/L TP obtained from the literature.

To speciate phosphorus, PP and DOP were computed from TDP, TP and OP parameters using Eqs. (6) and (7). The percentage composition of the phosphorus species indicated that approximately 51% of the total phosphorus was particulate bound and 49% was present in dissolved form. In the dissolved fraction, 19% of TP was present as dissolved inorganic phosphorus (OP) and 30% as DOP (Fig. 3).

The predefined EPA numerical criterion across all the 14 ecoregions for lakes and reservoirs for TN is 0.1–1.27 mg/L and for TP is 0.008–0.038 mg/L; for rivers and streams the TN criterion is 0.12–2.18 mg/L and TP is 0.001–0.128 mg/L (US EPA, 2020). The stormwater geomean concentrations for TN and TP exceeded all but the least stringent value for TN, confirming nitrogen and phosphorus as priority pollutants in urban stormwater.

Table 5
Summary statistics of Stormwater EMCs of nutrients from the International BMP Database and the National Stormwater Quality Database.

Water quality constituents	Units	N	%BDL	Min.	Median	Mean	Max.	Geomean	GSD
NH_4-N	mg/L	4142	15.7	0.003	0.27	0.61	53	0.25	3.95
NO_3-N	mg/L	2141	6.1	0.0002	0.5	0.85	52.1	0.48	2.94
NO_2-N	mg/L	917	39.0	<0.001	0.044	0.10	2.49	0.05	3.26
NO_2-N + NO_3-N	mg/L	4360	2.4	0.001	0.497	0.70	13.7	0.44	2.84
TON	mg/L	693	0	0.02	0.55	0.88	19.4	0.56	2.42
TKN	mg/L	6271	2.4	0.001	1.31	1.91	66.4	1.32	2.34
TN	mg/L	2186	1.7	0.05	1.47	2.08	53.3	1.5	2.21
OP	mg/L	1835	17.5	<0.001	0.03	0.15	83.2	0.03	5.41
TDP	mg/L	3443	12.5	0.001	0.1	0.18	19.3	0.09	3.2
TP	mg/L	7961	3.4	0.002	0.21	0.35	28.9	0.2	2.81

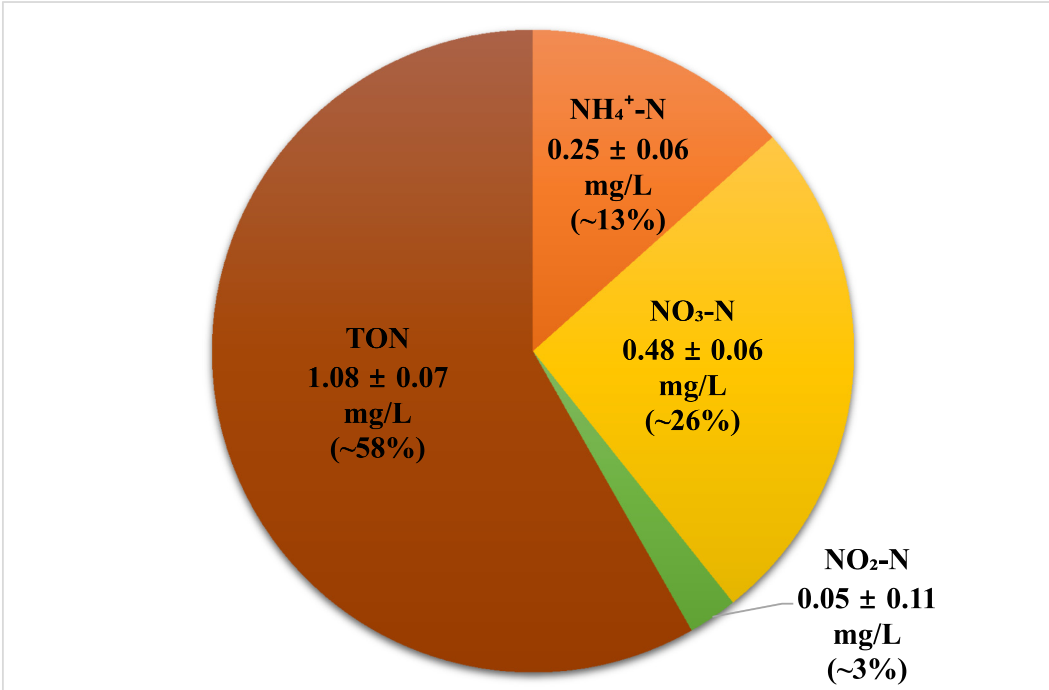

Fig. 2. Percentage breakdown of nitrogen species based on geomean values (TN = 1.85 ± 0.13 mg/L as N); TON and TN are calculated using mass balances from other species – see text for details, format = geomean ± GSE.

Atmospheric deposition, human and animal waste, reclaimed water irrigation, fertilizer applications, detergents, and plant and leaf litter are among the sources that contribute to nonpoint source nutrient pollution (US EPA, 2016). Nutrient-enriched surface waterways lead to eutrophication of receiving waterbodies, resulting in damage to the aquatic ecosystem, and eventually loss of biodiversity (US EPA, 2016).

3.4. Metals

Trace elements chosen from the databases for preliminary water quality analysis were arsenic (As), antimony (Sb), beryllium (Be), barium (Ba), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), selenium (Se), silver (Ag), and zinc (Zn). Although As is a metalloid, it was included under the trace metals category for this study. Total as well as some of dissolved fractions are recorded in the databases. Of the available values, any metals with >50% of BDL were omitted from water quality analysis since the data become unreliable for any estimation methods. The ROS method was adopted for estimating non-detects for all the trace metals because of the significant non-detect populations. Table 6 provides summary values of the chosen metal constituents from the databases. The geomean concentrations for the dissolved metal species were less than or equal to their total metal values, as it is required to match mass balances (total > dissolved).

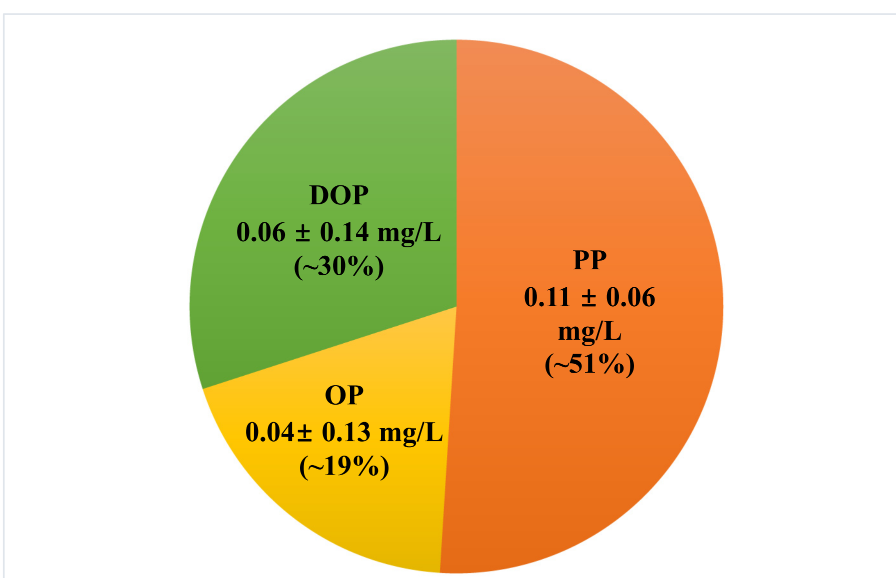

Fig. 3. Percentage breakdown of phosphorus species based on geomean values ± GSE (TP = 0.2 ± 0.03 mg/L as P). At pH 7.1, the OP is speciated at 56% $H_2PO_4^-$ and 44% HPO_4^{2-}.

The metals data were right skewed, with most of the datasets not following a normal distribution even after log transformations, owing to significant non-detects and potential outliers. Among metals, Fe clearly dominated and occurred at a wider range than any others (Table 6). Data that were compiled from peer-reviewed literature on metals as opposed to the databases, most commonly analyzed for Zn, Cu, Cd, Cr, and Pb, with fewer studies testing for Fe, as Fe has much lower toxicity than the others.

The particulate fractions of Cu, Cr, Ni and Zn were calculated from the geomean dissolved and total metal concentrations to estimate metal partitioning. The metal(loid)s As and Fe were eliminated from the partitioning analysis, since the former will require an assumption of oxidation state, and the latter is ubiquitous and not a typical pollutant of concern. Previous studies emphasized the importance of metal partitioning as most of the stormwater heavy metals are particulate-bound (Gnecco et al., 2008, 2019; Herngren et al., 2005; Shinya et al., 2000; Zgheib et al., 2011). As seen in Table 7**,** Cr, Ni and Zn were predominantly particulate-bound, whereas Cu was mainly present in the dissolved phase. Gnecco et al. (2019) found that Cu was particulate-bound and that the Zn partitioning behavior changed as a function of monitoring site. Shinya et al. (2000) and Zgheib et al. (2011) also observed similar Cu partitioning, with Cu exhibiting greater affinity for the particulate phase. However, Herngren et al. (2005) pointed out that the Cu partitioning varied from site to site, and Cr strongly correlated with TSS across sites based on GAIA (Geometric Analysis for Interactive Aid) analysis. Zgheib et al. (2011) also reported 52% of Zn to be particulate and less correlated with the suspended solids among the metals analyzed in the study. This difference in metals partitioning trends across studies brings forth some of the contributing factors which include land use, stormwater solution parameters, suspended solids load, particle size distribution, and runoff volume. A few studies also evaluated physicochemical characterization of the solids for more in-depth and detailed analysis of the solid phases as PM has surface chemical functional groups that will bind metals. The geomean values of total and dissolved metals are distinct because the data are derived from different populations, and they do not necessarily depend on each other. However, this establishes a certain knowledge of metal partitioning.

Establishing aqueous speciation of metals in stormwater and their partitioning between dissolved and particulate phases is of importance for optimizing treatment and evaluating the environmental impacts of stormwater. For example, soluble ionic forms of metals or free metal ions are more toxic to aquatic life than particle-bound or complexed species. Therefore, understanding the speciation of metal ions can fundamentally contribute to establishing the design traits of a stormwater SCM.

Metal speciation is a strong function of pH, the presence of PM, and inorganic and organic ligands. Due to limited data availability on PM characterization and the complexity of metal/PM surface interactions, metal speciation and respective compositions were determined using the aqueous concentrations of the metals. Geomean values of conventional water quality aggregates values of pH, inorganic (Cl^-, PO_4(-III), NO_3^-) and organic (DOC) ligands concentrations, alkalinity, and conductivity (for calculating ionic strength), along with dissolved metals concentrations, were used as inputs in Visual MINTEQ 3.1 (Table 8**).** DOC/TOC values are chosen as surrogate measures to determine the organic matter content in stormwater (Barrett et al., 2014).

Results from speciation modelling (Fig. 4) showed that all the dissolved metal ions under typical stormwater conditions were complexed to DOM either electrostatically (weak complex: Me–HA), or through inner sphere complexes (strong) with carboxylic (MeHA1) or phenolic sites (MeHA2) of humic acid (HA). Changing the NICA-Donnan model parameters such as ratio of active DOM to DOC and percent of active DOM that is fulvic/humic can change the binding properties of DOM with the metal ions. In this study, the percent of inner sphere complexes among the metals followed the order Cr(III) > Cu(II) > Ni(II) > Zn(II). From Fig. 4 it can be observed that Ni(II) and Zn(II) form rather weak HA complexes compared to Cr(III) and Cu(II). Using GAIA analysis, Herngren et al. (2005) also observed that Cu(II) and Zn(II) displayed a high correlation with dissolved organic carbon. Similarly, other studies concluded that the presence of DOM can change the preference of complexation to DOM as opposed to carbonates or other ligands (Dean et al., 2005; Gnecco et al., 2008). This underscores that DOM is a critical stormwater quality parameter. Phosphate and carbonate ligands had no effect on metals speciation under the given input conditions.

Stormwater is responsible for a substantial washout of trace metals into receiving water bodies (US EPA, 2015b). Trace metals have been prioritized in terms of pollution control for years because of their persistence and acute toxicity, despite their occurrence at relatively low concentrations (ng/L to μg/L, Tchounwou et al., 2012; Sakson et al., 2018). Major metals sources include road asphalt, automobile brakes and tire wear, vehicle exhaust, oil spills, parking dust, building siding, metal-coated roofs, and airborne deposition (Davis et al., 2001; Reddy et al., 2014; Sansalone and Buchberger, 1997b). De-icing salts may also contribute to elevated metal concentrations, particularly trace metals such a nickel and chromium (Thomson et al., 1994). Metal pollution loads depends on land use, rainfall intensity, antecedent dry period and traffic congestion (Herngren et al., 2005; Kayhanian et al., 2003, 2012).

Table 7
Partitioning of Cu and Zn at geomean concentration ± geometric standard error (percent composition). Particulate fraction is calculated as the difference between the geomean EMCs of total and dissolved.

Trace metal	Total (μg/L)	Dissolved (μg/L)	Particulate (μg/L)
Cu(II)	13 ± 0.05	7.5 ± 0.08 (57.3%)	5.6 ± 0.09 (42.7%)
Cr(III)	3.7 ± 0.05	1.4 ± 0.07 (37.6%)	2.3 ± 0.09 (62.4%)
Ni(II)	4.3 ± 0.05	1.8 ± 0.07 (42.7%)	2.4 ± 0.09 (57.3%)
Zn(II)	71 ± 0.04	30.3 ± 0.07 (42.8%)	40.5 ± 0.08 (57.2%)

Table 6
Summary statistics of stormwater EMCs of metal(oid) concentrations from the International BMP Database and the National Stormwater Quality Database.

Metal forms	Water quality constituents	Units	N	%BDL	Min.	Median	Mean	Max.	Geomean	GSD
Total	As	μg/L	2120	50.9	0.05	1.1	2.3	91	1.1	3.0
	Ba	μg/L	388	29.9	2	21.8	34.2	1520	20.2	2.6
	Cd	μg/L	4443	48.5	0.009	0.34	1	275	0.3	4.0
	Cr	μg/L	2321	31.9	0.2	3.9	6.1	559	3.7	2.6
	Cu	μg/L	5933	8.5	0.1	13	45.3	7270	13	3.6
	Fe	μg/L	800	8.4	9	650	1967	177,000	650	4.3
	Ni	μg/L	2184	30.6	0.003	4.4	6.6	281	4.3	2.5
	Pb	μg/L	5089	21.7	0.075	7.3	22.3	2300	7	4.9
	Zn	μg/L	6490	3.8	0.06	75.7	144	27,500	71	3.3
Dissolved	As	μg/L	1307	48.8	0.05	0.7	1.7	67	0.7	3.2
	Cr	μg/L	1673	44.7	0.05	1.4	2.4	41	1.4	2.9
	Cu	μg/L	3058	8.8	0.3	6.5	51.6	3540	7.5	4.3
	Fe	μg/L	834	40.2	0.1	50	152.4	7600	52.4	3.9
	Ni	μg/L	1664	43	0.002	2	3.1	82	1.8	2.9
	Zn	μg/L	2921	8.3	0.1	29	76.4	25,800	30.3	3.6

Table 8
Input values (geomean) required for metals speciation.

Input parameter	Input value
pH	7.1
Temperature (°C)	15
Conductivity (μS/cm)	97
Ionic Strength (M)[a]	0.0016
Alkalinity (mg/L as $CaCO_3$)	22.5
Cl^- (mg/L)	7.3
DOM as DOC (mg/L)	10.3
OP as P (mg/L)	0.04
Cu_d (μg/L)	7.5
Cr_d (μg/L)	1.4
Ni_d (μg/L)	1.8
Zn_d (μg/L)	30.3

[a] Ionic Strength calculated from conductivity (Eq. (1)).

Dissolved fractions of metals are more bioavailable and detrimental to marine organisms than particulate fractions (LeFevre et al., 2015). Therefore, it is imperative to target dissolved metal fractions to attenuate their impacts on water quality and aquatic life. PM and dissolved solids in stormwater are strongly associated with heavy metals and have an influence on metal partitioning and speciation behavior (Dean et al., 2005; Gunawardana et al., 2012; Herngren et al., 2005). Gunawardana et al. (2012) showed that heavy metal loadings in PM increase as the particle size decreases. Similar observations for metals have also been noted by past researchers, and this occurs due to the large surface area and higher cation exchange capacity (CEC) of the finer sediments (Charlesworth and Lees, 1999; Herngren et al., 2005).

3.5. Organic pollutants

3.5.1. Polycyclic Aromatic Hydrocarbons (PAHs)

The databases contain information on organic pollutants including polycyclic aromatic hydrocarbons (PAHs), pesticides, and a few volatile organic compounds. Information pertaining to PAHs was well-reported in the databases, where individual and combined PAHs (total, dissolved, and suspended) accounted for 3074 data points. Concentrations of individual PAHs with over 100 data points and less than 50% BDLs were obtained from the databases (with one exception, described below). Suspended and dissolved PAH concentrations had fewer datapoints (<30) and not all individual PAHs were represented in the databases. This limited the analysis to the data of total (dissolved + particulate) concentrations and made it impossible to assess the partitioning behavior of these pollutants. The

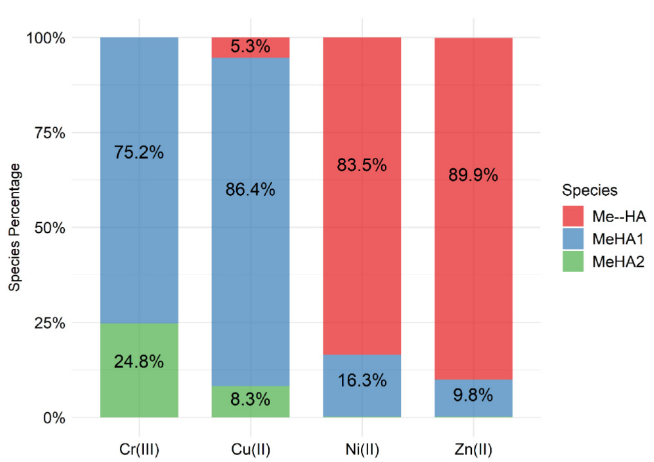

Fig. 4. Speciation of dissolved Cu, Cr, Ni and Zn at fixed stormwater conditions (input parameters are listed in Table 8). All metals are dominated by DOM (humic acid, HA) complexation: electrostatic weak complex: Me–HA, strong inner sphere complex with carboxylic sites: MeHA1, strong inner sphere complex with phenolic sites: MeHA2.

robust ROS method estimated the concentrations of BDLs for all the total PAHs. Table 9 presents the EMC summary statistics of 11 of 16 priority PAHs that were chosen for data analysis.

The summation of geomean EMCs for individual PAHs yielded a value of 1.65 μg/L, which is close to the median value of ΣPAHs = 1.33 μg/L measured in a study conducted in Paris (Zgheib et al., 2012) and 25 times higher than the mean value of ΣPAHs = 0.067 ± 0.028 μg/L found in the Susquehanna River Basin in Maryland, USA (Ko and Baker (2004). This value also falls within the range of 1.5–12.5 μg/L from a storm flow reported in a study of the Tidal Anacostia River in the District of Columbia and Maryland (Hwang and Foster, 2006). Masoner et al. (2019) noted ΣPAH EMCs greater than 10 μg/L at many of their sites in a USA-wide study, with a maximum concentration of 180 μg/L. It appears that PAH concentrations vary significantly based on the characteristics of the drainage area and whether the PAH sources are pyrogenic.

The maximum concentrations of FLN, NAP, PYN and PHN were recorded at 22.8, 14.7, 15.8, and 40.4 mg/L respectively, and measured at a wider concentration range than other PAHs (Table 9). PAH values that were reported >1 mg/L in the databases were sampled from one low-density residential site in California. A study conducted in a parking lot in Maryland assessed the distribution of stormwater total PAHs (dissolved + particulate) and found that FLN and PYN were found at higher concentrations than other PAHs (DiBlasi et al., 2009).

Examining the geomeans of individual PAHs, BbFN was observed at the highest concentration, with PYN, BkFN, and FLN following in that order (Fig. 5). Only 77 data points were found in the database for BbFN, but since the geomean was the highest of the PAH, more than two times that of PYN and three times that of BkFN, it was included in the analysis (geomean = 0.69 μg/L). A Wisconsin study measured concentrations of 18 PAHs from six urban areas and observed FLN, PYN, CHR and BbFN to be the dominant PAHs (Selbig, 2009). Masoner et al. (2019) indicated the following order of PAHs: BbFN > FLN > PYN > CHR > BkFN, all at median concentrations around 0.5 to 1.1 μg/L. Cumulatively, it was concluded that the PAHs BbFN, BkFN, CHR, FLN, and PYN can be expected to make up a large proportion of total PAH concentration in stormwater.

3.5.2. Previous research on organic pollutants in stormwater

Literature was mined for PCBs concentrations in stormwater as the databases did not contain information pertinent to PCBs. The concentrations of PCBs varied across different land covers, with some studies reporting values lower than detection limit and some at higher concentrations. Zgheib et al. (2012) found ΣPCBs concentrations from 0.01 μg/L to 0.727 μg/L, with a median value of 0.25 μg/L. Hwang and Foster (2008) reported a similar range for ΣPCBs from 0.01 to 0.211 μg/L. However, a study from San Francisco, CA showed lower concentrations at 0.18–1.3 ng/L (Nicole et al., 2015). Since most studies presented a range of PCBs concentrations or solely a mean/median concentration, statistical analysis was not performed on the collected data.

To highlight the importance of other anthropogenic organic contaminants in stormwater, a nationwide study of 438 organic chemicals (e.g., biogenic hormones, halogenated chemicals, household/industrial chemicals, methylmercury, pesticides, pharmaceuticals, and semivolatile compounds) was recently completed (Masoner et al., 2019). This study evaluated results from 50 storm events at 21 sites across the U.S. to understand the contribution of these pollutants to natural water bodies. Half of the of 428 target chemicals were detectable, of which PAHs and pesticides were found frequently with 19% and 35% of total detections respectively. Even though % total detections was higher for pesticides than PAHs, concentrations of ΣPAHs were up to 180 μg/L (from coal-tar based sealcoats) and pesticides were detected from <0.001 μg/L to >10 μg/L. Additionally, caffeine and nicotine were the most frequently detected nonprescription pharmaceuticals at maximum concentrations of 32.3 μg/L (median = 0.942 μg/L) and 18.3 μg/L (median = 0.782 μg/L), respectively. Total PCBs were found less frequently (the only frequently detected congener being PCB 138 at 18% total detections), and concentrations varied from 2 to 84 ng/L.

Table 9
Summary statistics of Stormwater EMCs of Total PAHs from the International BMP Database and the National Stormwater Quality Database.

Water quality constituents	Units	N	%BDL	Min.	Median	Mean	Max.	Geomean	GSD
Benz[a]anthracene (BaA)	μg/L	179	43	0.002	0.01	0.26	7	0.02	13.1
Benzo[b]fluoranthene (BbFN)	μg/L	77	23.4	0.13	0.71	1.69	18	0.69	3.9
Benzo[a]pyrene (BaP)	μg/L	173	44.5	0.004	0.02	0.49	12	0.03	15.3
Benzo[ghi]perylene (BghiP)	μg/L	176	27.8	0.003	0.04	0.58	14	0.04	14.4
Benzo[k]fluoranthene (BkFN)	μg/L	110	19.1	0.012	0.21	0.58	8.2	0.22	3.8
Chrysene (CHR)	μg/L	180	29.4	0.003	0.05	0.69	17	0.05	15
Fluoranthene (FLN)	μg/L	183	12	0.003	0.11	535	22,800	0.17	50.7
Indeno[1,2,3-cd]pyrene (IPY)	μg/L	172	32	0.003	0.02	0.56	13	0.03	18.2
Naphthalene (NAP)	μg/L	188	44.7	0.005	0.01	498	14,700	0.02	42.5
Phenanthrene (PHN)	μg/L	184	12.5	0.003	0.06	463	15,800	0.14	42.7
Pyrene (PYN)	μg/L	181	9.4	0.003	0.17	880	40,400	0.24	46.5

Tran et al. (2019) conducted a study in Singapore, examining 31 emerging contaminants (ECs) in urban and agricultural stormwater runoff. In 51 samples from this study, 17 of 31 target ECs were detected. The most frequently detected compounds (100% frequency) in urban stormwater belonged to: artificial sweeteners, industrial chemicals (bisphenol A), neuroactive drugs, caffeine, the pesticide DEET, nonsteroidal anti-inflammatory drugs, and antiseptic additives. Some ECs were frequently detected at a median concentration > 100 ng/L (artificial sweeteners, nonsteroidal anti-inflammatory drugs, bisphenol A, caffeine, DEET) while others were detected with median concentrations ≤ 10 ng/L.

Examination of 69 organic pollutants including pharmaceuticals, herbicides, industrial chemicals, and PAHs from stormwater in combined sewer overflows (CSOs) was performed in Stuttgart, Germany (Launay et al., 2016). The results showed found that herbicides, PAHs, and one industrial phenol compound were major contributors, with the group of urban herbicides being highest ranging from 93% to 99%. It was also found that stormwater contributed ~50% of the load of two other industrial chemicals and ~ 30% of the neuroactive drug compound load (compared to wastewater discharges). Launay et al. (2016) concluded that stormwater was the most important source for all herbicides, 4tOP (an industrial phenol compound), and PAHs.

Mei et al. (2018) examined pharmaceutical and personal care products (PPCPs) in surface water samples from the Huangpu River in Shanghai, China and compared wet weather and dry weather samples to evaluate the contribution of PPCPs from stormwater. The results showed that in rural areas the concentration of PPCPs decreased during storm events, likely due to dilution in receiving water and clean runoff from rural areas. In contrast, the authors found that PPCPs concentrations increased in urban areas after rainfall event, especially for caffeine and carbamazepine (CBZ, a neuroactive drug compound).

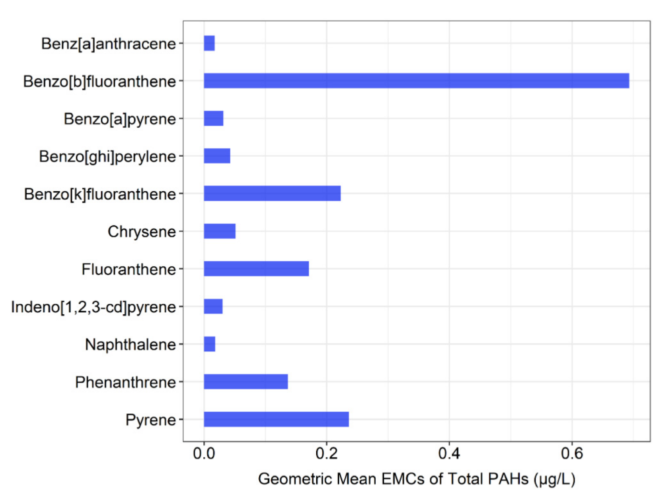

Fig. 5. Distribution of specific PAH compounds representing Total PAHs in stormwater based on geomean of the event mean concentrations.

Zgheib et al. (2011) evaluated stormwater from six storm events at an urban catchment in Paris, France. Particulate and dissolved phase occurrence and concentrations were examined for 88 stormwater priority substances, of which 45 were quantified in the 6 samples collected in this study, and 37 were recurrent (observed in 75%–100% of samples). The quantified substances included 4 metals, 1 volatile organic compound, 7 PCB congeners, 16 PAH compounds, 3 organotins, 1 chlorophenol, 5 alkylphenols, 1 phthalate, and 7 pesticides. Most of these substances partitioned predominantly to the particulate phase (Zgheib et al., 2011).

Burant et al. (2018) conducted a study on aqueous phase (<1.6 μm) trace organic contaminants in urban runoff at a commercial and residential site in Wisconsin, USA (2018). Here, 81 contaminants were evaluated including pesticides, flame retardants, PAHs, corrosion inhibitors, and PPCPs and others. Of the targeted compounds, 30 had >50% detections for at least one of the sites. Detected PAHs were present within higher traffic areas, which can be explained by presence of combustion products, and temporal trends with pesticide application, but not significant difference between land-use. Contrary to the hypothesis in this study, residential sites showed higher total concentrations of flame retardants and corrosion inhibitors than the commercial site (Burant et al., 2018).

Many persistent organic pollutants are toxic and can pose detrimental effects to aquatic and human life, given their tendency to bioaccumulate and biomagnify within the food chain. These pollutants are produced as a result of agriculture, industrial activities, combustion, transportation, urban landscape management, and various other human activities. Many of these organic chemicals are capable of long-term transport and are densely found in urban areas.

3.6. Pathogens and indicators

Information about Fecal Indicator Bacteria (FIB) was retrieved from the databases including *Escherichia coli* (*E. coli*), Fecal Coliform (FC) and the Fecal Streptococci group (FS). Bacterial abundance was monitored using Most Probable Number (MPN) showing the number of viable cells per 100 mL of the sample. Additionally, geomean was excluded from summary statistics because some datapoints were reported at 0 MPN/100 mL, which would yield a non-zero value when calculated; median was used as measure to identify typical bacterial concentrations in stormwater.

Table 10 presents the summary statistics of EMCs of pathogens/indicators. FIBs had the widest range of abundance among the stormwater pollutants: concentrations of *E. coli*, FC and FS ranged from 0 to 1.7×10^7 (median = 990) MPN/100 mL, 1 to 6.0×10^6 (median = 5000) MPN/100 mL, and 2 to 1.8×10^6 (median = 3×10^4) MPN/100 mL, respectively (Table 10). Paule-Mercado et al. (2016) estimated the abundance of *E. coli* and FS as $10^{2.51}$ to $10^{7.39}$ and $10^{2.48}$ to $10^{7.21}$ MPN/100 mL respectively. This range corroborated with the FS median of 3×10^4 MPN/100 mL of the current study, but not that of *E. coli*. A study conducted in Australia monitored *E. coli* abundance across various catchments with a range of 4.7×10^2 to 1.82×10^4 MPN/100 mL under wet weather conditions (Sidhu et al., 2012) which support the data listed in this study.

Pan and Jones (2012) examined statistical variations in FIB abundance using data from the BMP database, observing that fecal bacterial numbers

Table 10
Summary statistics of stormwater EMCs of pathogens concentrations from the International BMP Database and the National Stormwater Quality Database.

Water quality constituents	Units	N	%BDL	Min.	Median	Mean	Max.	SD
E. coli	MPN/100 mL	384	3.1	0	900	1.9×10^5	1.7×10^7	1.3×10^6
FC	MPN/100 mL	1525	1.1	0.5	5000	7.6×10^4	6×10^6	4.3×10^5
FS	MPN/100 mL	599	1.2	1	3×10^4	7.5×10^4	1.8×10^6	1.5×10^5

depended on the season, with a potential for higher bacterial flux during summers. Similarly, a field study in South Korea found an inter-summer variability of FIB concentrations, correlating higher concentrations to higher temperatures (Paule-Mercado et al., 2016). Similar to other pollutants, bacteria can also be associated with settleable particulate matter (Krometis et al., 2009; Sidhu et al., 2012). In general, seasonal changes, catchment type, rainfall intensity were some of the factors that have been noted to affect the EMCs of pathogens (Blaustein et al., 2016; Pan and Jones, 2012; Paule-Mercado et al., 2016; Sidhu et al., 2012).

Microbial species, which are traditional indicators of waterborne disease contamination, are also monitored in stormwater. From a public health vantage point, the importance of treating/reducing pathogens from urban stormwater is accented when discharge waterbodies are used for drinking water supply, swimming, or other recreational activities. Deposition of fecal waste from animals and humans appear to be potential sources of bacteria in stormwater (Ahmed et al., 2019; Steele et al., 2018; US EPA, 2002).

4. Conclusions

The International Best Management Practices database and National Stormwater Quality database were utilized to characterize urban stormwater and categorize its water chemistry and pollutants. Urban stormwater characterization included contaminants of concern as well as ubiquitous background constituents affecting pollutant speciation and partitioning. To supplement the database data, a literature review was performed on particle size distributions of stormwater particulate matter and emerging pollutants. In the combined databases, stormwater constituents with greater detectable concentrations followed a log-normal distribution, while the BDL-laden datasets did not.

Mass balances based on the geometric mean of EMCs for nitrogen species were not met when totals (TN, TKN, TON) were compared to the sum of the individual species (NH_4-N, NO_3-N, NO_2-N) because the data were derived from discrete datasets. A more reliable statistical analysis could be achieved if these datasets from the same study/storm event were grouped together, providing complete speciation information for a specific pollutant. Nutrient speciation indicated that a significant portion of phosphorus was particulate-bound, whereas nitrogen was present in the dissolved form.

Literature review suggested that trace metals partitioning is dependent on the catchment site/characteristics and traditional water quality constituents, while the statistics on EMCs from the databases showed that Cr, Ni and Zn were most abundantly detected in the particulate phase, while Cu was most often present in the dissolved phase. Notably, DOM was shown to impact metals speciation, with metals entirely complexed with DOM at pH ~7.

The combined databases carried rich data for PAHs compared to other organic chemicals. Between observations from the databases and literature studies, it can be concluded that the total PAH is mostly composed of BbFN, BkFN, CHR, FLN, and PYN. Recent research has found PAHs, PCBs, a suite of emerging contaminants like PPCPs, herbicides, and other synthetic chemicals present in stormwater from urban regions.

5. Research needs

Public databases (such as BMP and NSQD) of stormwater studies are valuable sources for information on stormwater and its management. The databases principally contain studies from the U.S. with limited data from other countries. While maintaining such large databases is a large effort, the databases are a wealth of information and they must be continuously updated, more stormwater studies must be included from other countries/continents, and emerging environmental contaminants should be included to provide valuable information for stormwater practitioners and researchers.

Stormwater characterization is a multivariate analysis and is dependent on land uses and activities, seasons, climate, antecedent dry periods, and rainfall intensities. Thus, greater understanding of stormwater chemistry and biology from other geographic areas and climate zones will expand the understanding of stormwater risks and SCM performance information to these areas.

Fundamental work on characterizing stormwater chemistry and microbiology is needed to provide greater understanding of environmental impacts and to improve SCM designs. The most challenging aspects of the characterization are the organic fractions, which contribute to organic N and P, and control speciation for several important trace metals. Similarly, the PM fraction is important as a pollutant but is also a carrier of many traditional and emerging pollutants. Complete speciation information is needed for nitrogen and other parameters to close mass balances and to design processes to target these species. Characterizing the complex structural properties (humic, fulvic and inert fractions) of stormwater dissolved organic matter (natural and anthropogenic) and identifying its sources based on land uses is critical to evaluate and accurately predict metal speciation. More information is needed on identifying and quantifying trace-level metals and organic chemicals, as most of these constituents in the databases were listed as below detection limits, and/or contained few data points. While such studies are expensive to conduct, this information is needed to appropriately evaluate environmental and ecological risks of urban stormwater.

CRediT authorship contribution statement

Sai Thejaswini Pamuru: Investigation, Data curation, Writing – original draft, Writing – review & editing. **Erica Forgione:** Investigation, Writing – original draft. **Kristen Croft:** Investigation, Writing – original draft. **Birthe V. Kjellerup:** Supervision, Writing – review & editing, Project administration, Funding acquisition. **Allen P. Davis:** Conceptualization, Supervision, Writing – review & editing, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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