Metals and bacteria partitioning to various size particles in Ballona Creek stormwater runoff

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Abstract

Based on the well documented association between stormwater particles and pollutants associated with urban runoff, many stormwater Best Management Practice devices (BMPs) function primarily by capturing particulate matter. The hydrodynamic separation or settling methods used by most BMPs are most effective at capturing medium to large particles; however, these may not be the most predominant size particles associated with urban runoff. This research examined particle size distribution in stormwater runoff from an urban watershed in southern California and investigated the pollutant-particle associations of metals (Cu, Pb, Ni, Zn) and bacteria (enterococci, *Escherichia coli*). During small storm events (≤0.3" rain), the highest concentration of pollutants were associated with a <6 µm filter fraction, which accounted for 70% of the per storm contaminant mass, but made up <20% of the total particle mass. The pollutant-particle association changed with storm size, as most pollutant mass was associated with >35 µm size particles during a 2" rain event. These results suggest that much of the contaminant load in stormwater runoff will not be captured by the most commonly used BMPs, since the majority of these devices (e.g., hydrodynamic separators) are unable to capture particles <75 µm.

Introduction

Pollutants in stormwater are associated mainly with suspended particles, which act as the transport vector to downstream areas (Sansalone and Buchberger 1997, Lau and Stenstrom 2005, Surbeck *et al*. 2006, Lau *et al*. 2009). Past studies of runoff from parking lot and road surfaces demonstrated the majority of pollutants are associated with fine particles typically less than 50 µm (Furumai *et al*. 2002, Vaze and Chiew 2004). These fine particles have a propensity to settle downstream and accumulate in bays and estuaries where they may contribute to sediment contamination (Bay *et al*. 1998, Jeng *et al*. 2005).

In many urban watersheds, such as the Ballona Creek watershed, pollutants in stormwater runoff and in downstream bays and estuaries have resulted in waterbodies being listed as impaired under Section 303(d) of the Clean Water Act. These waterbodies are then subject to Total Maximum Daily Load (TMDL) requirements to reduce loading of urban-associated contaminants such as metals, bacteria, and organic contaminants. The most common approaches to meeting TMDL requirements for urban stormwater are through installation of Best Management Practices (BMPs) designed to capture or treat pollutants before they are discharged to streams or receiving waters (Sample *et al*. 2003). The mechanism used by the majority of BMPs is settling or filtering of stormwater particles as a

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means of reducing loading of the attached pollutants (Ackerman and Stein 2008). Consequently, BMP effectiveness is a function of the efficiency of particle capture relative to the concentration of bound pollutants in the stormwater runoff.

The effectiveness of BMPs at pollutant removal is complicated by the fact that the timing of runoff of pollutants, including metals and bacteria during storms is not static. Pollutant concentrations and loads are typically higher during the early portion of storms and during the earlier storms of the season (Characklis and Wiesner 1997; Tiefenthaler *et al*. 2008, 2010). Although the majority of pollutants remain particle bound throughout most storms (Characklis and Wiesner, 1997), more recent research has shown that the ratio of particulate to dissolved concentration varies over the course of storms as a function of storm size, antecedent conditions, and surrounding land use (Yoon and Stein 2008). These factors can further affect BMP performance and concentration of pollutants delivered to downstream water bodies.

Management of stormwater runoff through BMPs can be improved by an increased understanding of the dynamic relationship between pollutants and particles. Because fine particles predominate in stormwater runoff, it is important to move beyond pollutant characterization as either particle-bound or dissolved. Knowing the relationship between pollutants and specific particle sizes over the course of storms, and between different storms within a season, will allow managers to more effectively target BMP design and implementation. Although these relationships have been described well in runoff from developed surfaces (Lau and Stenstrom 2005, Sansalone and Buchberger 1997), relatively less is known about how the partitioning between pollutants and various size particles changes over the course of storms and seasons sampled from urban flood control channels that integrate runoff from a variety of land surfaces.

The goal of this project was to improve the understanding of pollutant-particle relationships in urban stormwater runoff. The study focused on answering the following three questions based on data collected from the Ballona Creek Watershed: 1) What is the pollutant-particle association in urban stormwater? 2) Does the association change over the course of a storm? 3) Does the association change among storms?

Methods

Study Area

Sampling occurred in the Ballona Creek watershed, located in western Los Angeles County, California (Figure 1). The Ballona Creek watershed above the sampling point drains 230 km^2 and is approximately 85% developed, representing a typical urbanized watershed for southern California (Ackerman *et al*. 2005). Runoff from the upper half of the watershed is conveyed through a series of underground storm drains, discharging to a concrete-lined channel that ultimately flows to Santa Monica Bay. Sampling occurred from the Sepulveda Blvd. Bridge located 7.2 km downstream from where Ballona Creek becomes an open channel, 6.8 km upstream from the mouth, and 2.0 km upstream above the zone of tidal influence. Average annual rainfall in the watershed ranges from 340 to 530 mm, with the majority of storms occurring from January to March.

Sample Collection

Three types of measurements were made during a series of storm events over four storm seasons.

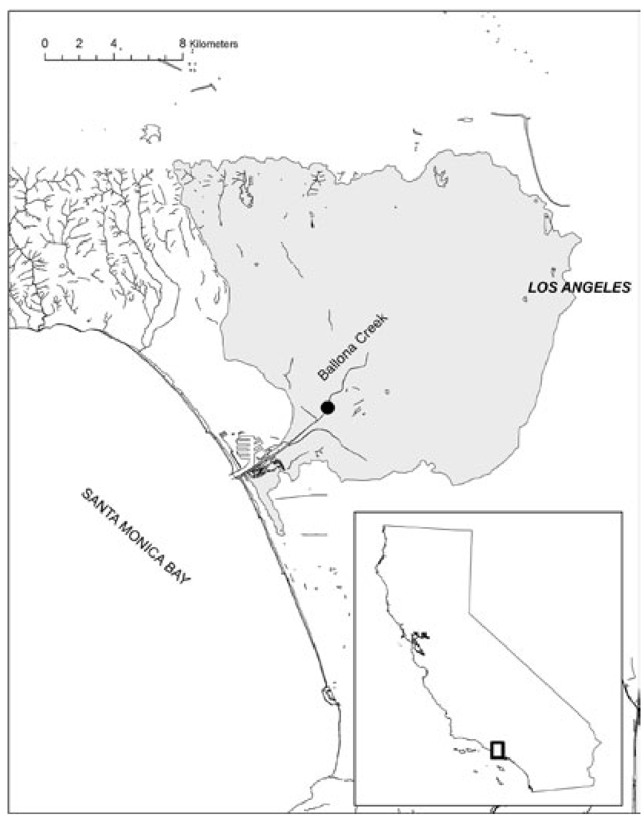

Figure 1. Sampling location within the Ballona Creek watershed in southern California. The shaded area represents the extent of the watershed.

Particle size distribution was measured over the course of six storms, trace metals were measured in four storms bound to various particle size fractions, and bacteria were measured in the particle bound phases during eight storms.

Samples for metals and bacteria were taken directly from the creek using a United States Geological Survey (USGS) depth-integrated sampler, or by collecting water pumped from the creek. For the pumped samples, stormwater from Ballona Creek was pumped vertically 10 m from the concrete channel bed to the top of the bridge over the channel. Water was pumped through Teflon® or silicon tubing (9.5 mm inside diameter) attached to a 25.4 mm angle iron bolted to the concrete channel bottom. Samples were pumped using a Masterflex I/P 77410-10 peristaltic pump with two heads in parallel and Masterflex Norprene I/P 73 tubing. Creek water was transported to the bridge and through a filter with 480 µm mesh sump filter (Cole-Parmer Low-cost In-line Strainer System). The bowl and mesh of the sump filter were manually switched out approximately every 15 minutes, or when they became clogged with leaves, small sticks, trash, etc. Pumping rates were maintained at >3 L/minute (0.8 m/s) to ensure that there was no particle settling in the tubing, based on validation studies conducted by Brown *et al*. (2009). Between three and ten samples were collected for each storm, with a greater number of samples collected during the earlier storms in this study. Samples were collected throughout each storm, often with a higher sampling frequency at the beginning of each event in order to characterize any first-flush response in metal or bacteria concentrations. Samples were held on ice and transported to the lab for filtration within one hour of collection to minimize flocculation and settling.

Particle size distribution was continuously monitored throughout most storms using the Laser In Situ Scattering and Transmissometry (LISST) device described by Brown *et al*. (2009). Laser refractometery produces particle density estimates by shining a laser through a parcel of water to calculate particle size distribution based on the amount of scatter induced by the particles. The method relies on assumptions about particle shape and density, and requires that water samples have sufficient transmissivity for penetration by the laser beam (Agrawal *et al*. 2008). Although this technique has not been widely used to measure particle size distribution in stormwater, Brown *et al*. (2009) demonstrated that with appropriate field procedures, *in situ* laser refractometry produces particle density and size distribution estimates that are comparable to laboratory methods.

Water was pumped from the creek, as described above, and run through the LISST to continually sample stormwater at 1 minute intervals. Technical difficulties with the pump setup prevented us from characterizing particle distribution for at least a portion of each storm. As such, we were only able to characterize the particle size distribution during the peak flow for six of the eight storm events sampled. Nevertheless, we characterized the distribution of particle sizes during the initial portion (i.e., rising limb of the hydrograph) of all storms, thereby allowing us to assess the portion of the hydrograph associated with first flush responses.

Flow data were obtained from the U.S. Army Corps of Engineers for the Ballona Creek flow gauge (http://www.spl.usace.army.mil/cgi-bin/cgiwrap/zinger/slLatestBasin.cgi?lacda+stage), located near the sample collection site.

Laboratory Analysis

Fecal indicator bacteria (FIB) and metals associated with various size fractions were measured by filtering stormwater through a series of progressively smaller filters (Table 1). Filter sizes ranged from 0.45 to 200 µm for the first storm, but because the measured distribution of metals and bacteria from early storms was mostly in the smaller fractions, the range was reduced to 6 and 35 µm for the last five storm events. Because of the difficulty in extracting bound particles from the filters, metals and bacteria were measured in the filtrate that passed through each filter. The constituent fraction associated with each size fraction was estimated as the difference in concentration between filtrate from successively smaller filters. Prior to filtering, concentrations were measured in each sample of raw runoff. The sample then was passed through the first filter in the series using a sterile 47 mm filtering funnel vacuum system. The filtrate was sampled using dilutions of 1, 0.1, and 0.01 ml for bacteria analysis, then passed through the next filter in the series.

Filtrate aliquots were tested for *Escherichia coli* using Colilert®-18, and for enterococci using Enterolert® (IDEXX, Westbrook, Me) (APHA *et al*. 1998: Standard Methods Section 9223 B). The concentrations of total metals in each filtrate were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) using EPA Method 200.8m. The metals analyzed included Cu, Pb, Ni and Zn.

Event flow-weighted mean concentrations (EMC) were calculated for the metals and bacteria data. Using only those samples for a single storm, the EMC was calculated according to Equation 1:

$$EMC = \frac{\sum_{i=1}^{n} C_i \bullet F_i}{\sum_{i=1}^{n} F_i} \qquad \text{Eq. 1}$$

where: *EMC* was event flow-weighted mean concentration for a particular storm; C_i was individual runoff sample concentration of i^{th} sample; F_i was instantaneous flow at the time of i^{th} sample; *n* was number of samples per event.

Data Analysis

In order to more readily compare data among the various filtration size classes used during different storms in this study, the metals and bacteria data were consolidated to three size ranges: <6 µm, 6 - 35 µm, and >35 µm.

Differences in the proportions of total particle mass represented by the different filter fractions were assessed using the Kruskal-Wallis analysis of variance on ranks, followed by the Student-Newman-Keuls multiple comparison procedure to identify which fractions were significantly different. Significance was determined at $\alpha = 0.05$.

Results

Particle Size Distribution

The dominant particle size varied over the course of each storm, with fine particles often dominating during the early part of the storm and the proportion of coarser particles increasing later in the storm. For most events, there was an increase in the silt/clay fraction (<63 µm, Wentworth scale; Wentworth 1922) as stormwater began to flow in the channel, with a peak in concentration that occurred before the peak in storm flow. The peak concentrations of the silt/clay fraction varied among storms by a factor of 23, ranging from 60 - 1356 µl/L. There was no significant relationship between the silt/clay peak concentration and preceding volume of stormwater ($p = 1.00$, Spearman rank correlation), flow velocity ($p = 1.00$), volume of stormwater before the peak in storm flow ($p = 0.95$), or number of dry antecedent days ($p = 0.35$). Because the silt/clay fraction was the predominant particle size class for the first 50 - 75% of the storm volumes, we

Table 1. Filter sizes (µm) used during the storms monitored for bacteria and metals bound to particulates. Because of the measured distribution of bacteria in the early storms, and the difficulty of passing stormwater through the smallest filter sizes, the range was reduced to 6 and 35 µm for the last storm events. "Raw" indicates samples that were not filtered before analysis.

Storm Date	Bacteria								Metals						
	0.45	1	5	6	35	125	200	Raw	1	5	6	35	125	200	Total
1/14/06	x	x	x		x	x	x	x							
3/3/06		x	x		x	x		x							
3/28/06		x	x		x	x		x							
2/3/08				x	x			x							
12/15/08				x	x			x	x	x		x	x	x	x
2/5/09				x	x			x	x	x		x	x	x	x
1/17/10				x	x			x			x	x			x
2/19/10				x	x			x			x	x			x

focused on this size class for the subsequent analysis of pollutant partitioning.

Distribution of Contaminants among Particle Sizes

Most trace metals, with the exception of Pb, were associated with fine particles. Cu, Ni, and Zn were most strongly associated with the <6 µm fraction (Figure 2). For example, 51% of the mean Cu EMC was associated with the <6 µm fraction, 33% associated with the >35 µm fraction, and 16% associated with the 6 - 35 µm fraction. In contrast, concentrations of Pb were slightly greater in the >35 µm filter size fraction (44% associated with the >35

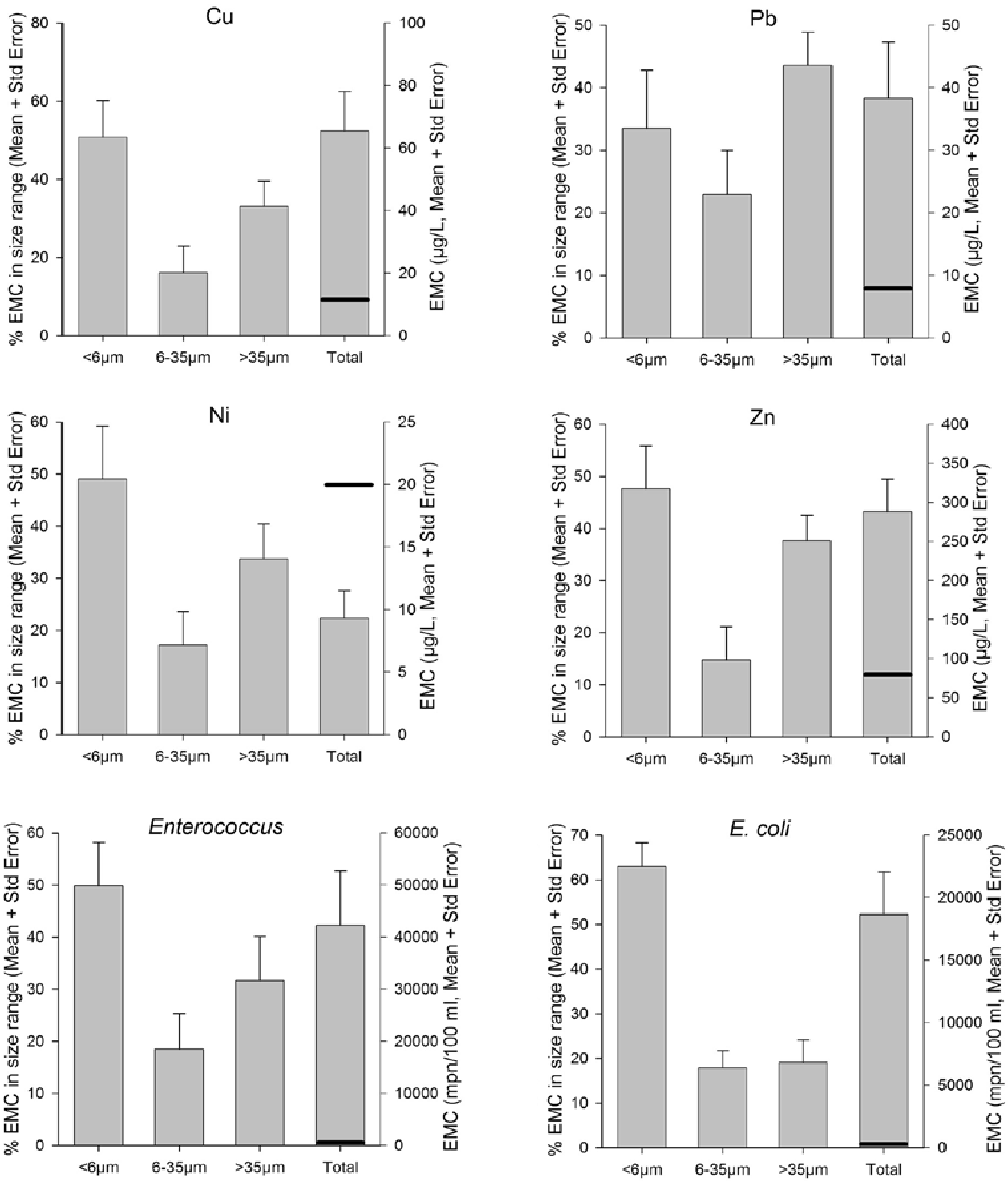

Figure 2. Proportion of metal flow-weighted event mean concentration associated with the various filter fractions (first 3 bars in each graph), and the overall EMC (fourth bar). The data represent the mean of the EMCs from the four (metals) or eight (bacteria) storm events. The horizontal slash on the fourth bar indicates the water quality standard used for comparison.

µm fraction, compared with 33% associated with the <6 µm fraction, and 23% associated with the 6-35 µm fraction).

Likewise, bacteria tended to be associated with the <6 µm fraction. Approximately 50% of the *Enterococcus* concentration and 63% of the *E. coli* concentration were associated with this smallest filter fraction (Figure 2). The EMCs varied; the proportion of enterococci associated with the <6 µm fraction ranged from 26 - 78% of the EMCs among the eight storm events, while the proportion of *E. coli* associated with this fraction ranged from 45 - 82%.

While the majority of the metals and bacteria concentrations were associated with the <6 µm fraction, this particle size fraction represented a significantly lower proportion of the total mass of stormwater particles ($H = 50.39$, $p<0.01$). Less than 25% of the total particle concentration was represented by the <6 µm particles (Figure 3). In contrast, the 6 - 35 µm fraction made up

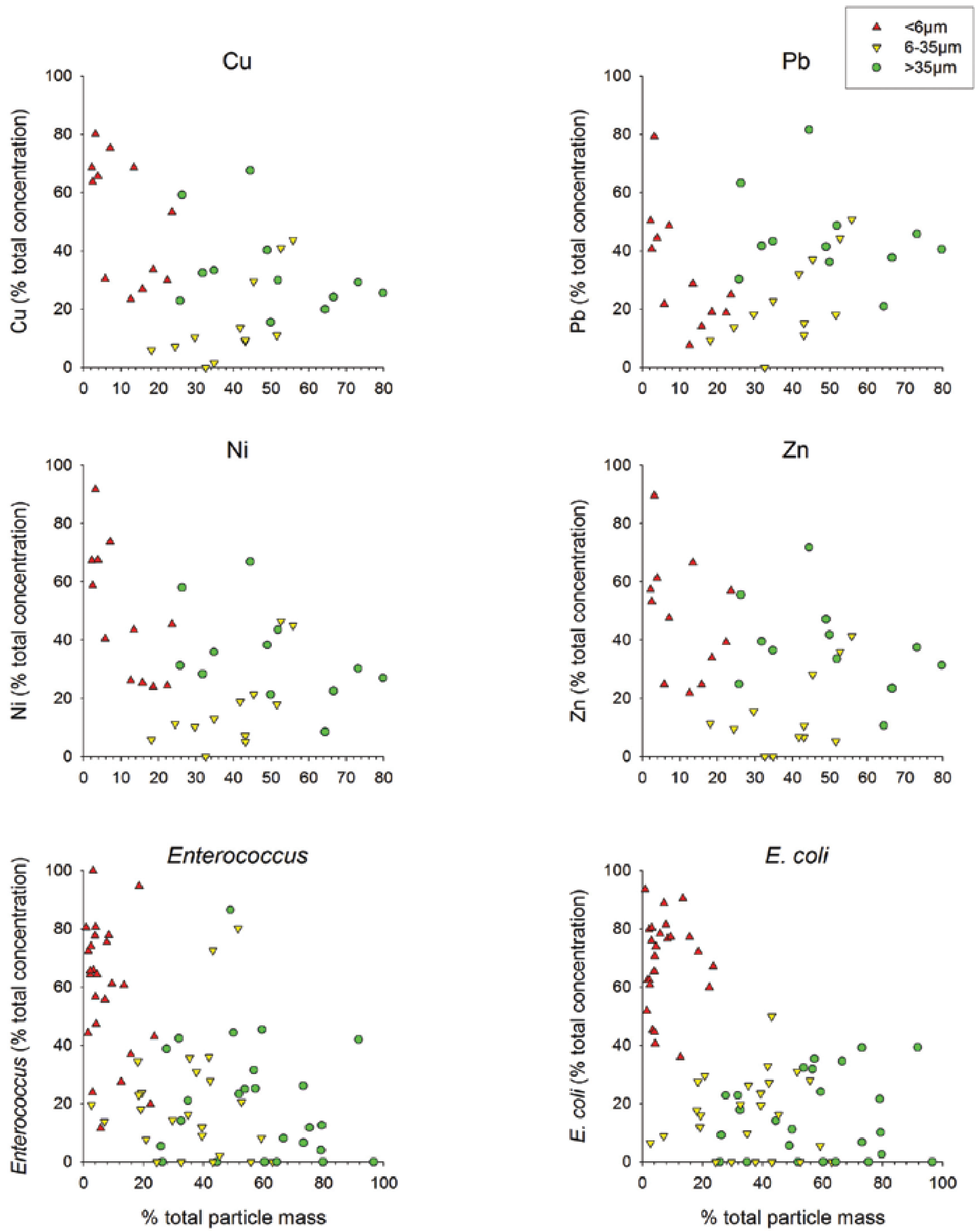

Figure 3. Association of metals and bacteria with the relative amounts of particles in stormwater samples. Data were pooled over four storm events for metals and eight events for bacteria.

approximately 39% of the total particle mass (on average), while the >35 µm fraction made up approximately 50%.

Within-Storm Variability

Metal concentrations were generally highest during the early portion of storms across all particle size classes. The highest concentrations typically occurred in samples collected before the peak in storm flow and decreased toward the recessional end of the hydrograph. The average ratio in concentrations of total metals between the first and last samples was 2.9 for Cu, 2.3 for Ni and 1.8 for Zn. For Pb, however, there was no difference in concentrations between the first and last samples collected. While concentrations of Cu, Ni and Zn tended to be highest at the beginning portions of storms, the peak in metal mass discharge coincided with the peak in flow rate for all particle size categories (Figure 4). This trend was consistent for each of the metals, including Pb.

Unlike metals, there was no consistent trend in bacteria concentrations over the course of the storms. Concentrations of enterococci and *E. coli* did not exhibit a first flush-like response, and bacteria concentrations appeared to be independent of stormwater flow volume at the time of sampling.

Like metals, the peak in bacteria (most probable number; mpn) discharge coincided with the peak in stormwater flow rate (Figure 4). This pattern was consistent for both enterococci and *E. coli*. The bacterial discharge associated with the individual fractions generally exhibited this same pattern.

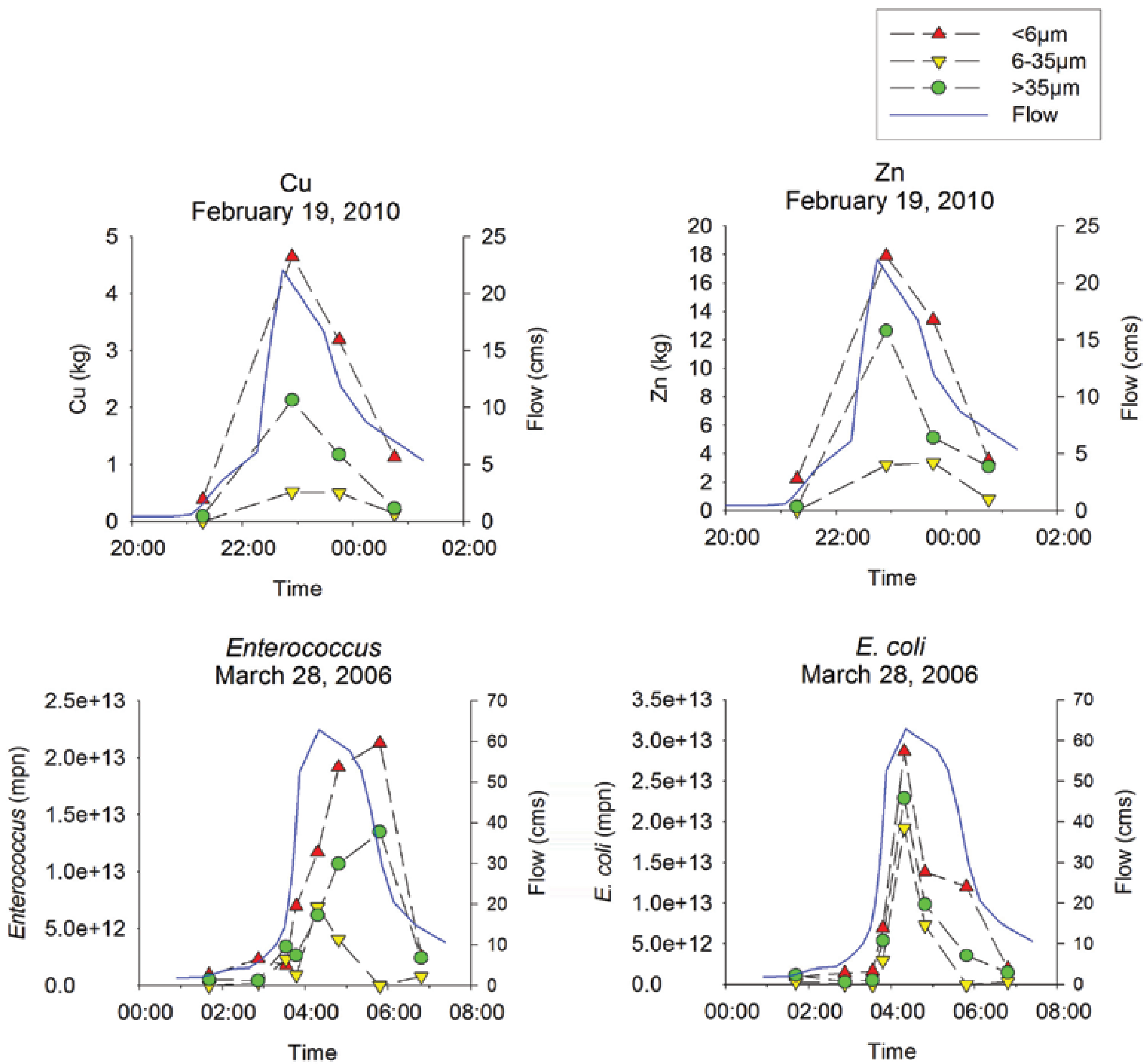

Figure 4. Metal and bacteria mass discharge distributed among the three filter fractions, relative to storm flow rate. The patterns for Cu and Zn mass discharge generally reflect those of Pb and Ni.

The pollutant-particle association was not always consistent within each storm. Cu mass discharge was consistently associated with the <6 µm fraction throughout storms with peak flow rates ≤50 cms, but alternated between the <6 µm, 6 - 35 µm, and >35 µm fractions for storms with peak flow rates >200 cms (Figure 4). Enterococci usually had a greater association with the <6 µm fraction throughout storms that had flow rates ≤60 cms, but this trend was not consistent (Figure 4); enterococci alternated between the <6 µm and >35 µm fractions over the course of the storm that had the lowest peak in flow rate (15 cms). *E. coli* discharge tended to have a consistently high association with the <6 µm fraction throughout the course of most events (Figure 4).

Among-Storm Variability

The association of metals with particle size appeared to be influenced by storm size. Storms with lower total flow volumes (≤4.7×10^5 m^3, ≤0.26" rain event) had relatively lower masses of metals discharged, and these metals had a greater association with the <6 µm filter fractions (Figure 5). At a storm volume of 1.7×10^6 m^3, the distribution of metals among the filter fractions was approximately equal. The largest storm in this study (4.0×10^6 m^3, 1.89" rain event) had the greatest metal mass discharge, and the metals tended to have a higher association with the >35 µm filter fraction.

Changes in the bacteria-particle association with storm size appeared to be indicator-dependent (Figure 5). Similar to metals, enterococci had a greater association with larger particle fractions as storm size increased. For storms <1×10^6 m^3, a greater proportion of the bacteria discharge was associated with the <6 µm fraction, but the association changed to the larger sized particles as storm size increased. In contrast, *E. coli* was associated with the <6 µm fraction throughout the range of storm sizes sampled.

Discussion

In the Ballona Creek watershed, most stormwater metals and bacteria were associated with the <6 µm filter fraction at lower storm volumes, but the association shifted to larger sized particles with larger storms. Stormwater volumes in Ballona Creek between 1987 and 1998 indicate 63% of the storms were small (≤700,000 m^3 daily flow volume), with only 6% of the events having flow volumes equal to or greater than the largest storms measured in this study (>4,000,000 m^3 daily flow volume). Therefore the majority of pollutants are expected to be associated with the <6 µm fraction for most events. Because larger storms discharge greater amounts of contaminants, however, the distribution of metals and enterococci among the particle size ranges is expected to be much more comparable on an annual mass discharge basis. Using the distribution of metals identified in this study with the storm volumes measured between 1987 and 1998, the mass of Cu discharge would have been very similar between the <6 µm (36% of the mass) and >35 µm size fractions (also 36% of the mass) over this time period. The distribution was similar for Ni (34% of the mass associated with the <6 µm fraction, 37% associated with the >35 µm fraction), but it is estimated that a greater mass discharge would have been associated with the >35 µm fraction for Pb and Zn. For Pb, 19% of the mass was estimated to be associated with the <6 µm fraction, compared to 44% associated with the >35 µm fraction. For Zn, the mass distribution was estimated to be 34% associated with the <6 µm fraction, and 42% associated with the >35 µm particles.

While the sampling location for this study was at the bottom of the watershed, and therefore included stormwater runoff from a mixture of land uses (residential, commercial, and transportation), the results in this study were similar to those from studies that focused solely on highway runoff, collected at the source of the runoff. For example, Li *et al*. (2005) observed that particles <10 µm make up less than 20% of the total particle mass in stormwater runoff from highways in southern California, which was similar to our observations. Li *et al*. (2005) also observed that the highest particle concentrations occurred within the first hour, and decreased rapidly thereafter, which is similar to the pattern measured in the runoff at Ballona Creek in this study. Sansalone *et al*. (1996) similarly observed that metal partitioning between particle and dissolved phase was related to rainfall and storm flow intensity in runoff from urban roadways. In our study of the runoff from an entire watershed, we observed that contaminant distribution among particle size was related to storm size, with greater association with larger particles during larger storms.

It is unclear if the majority of metals or bacteria were associated with particles, or if they were freely dissolved. Sansalone *et al*. (1996) determined that for metals in highway runoff, the majority of Cu,

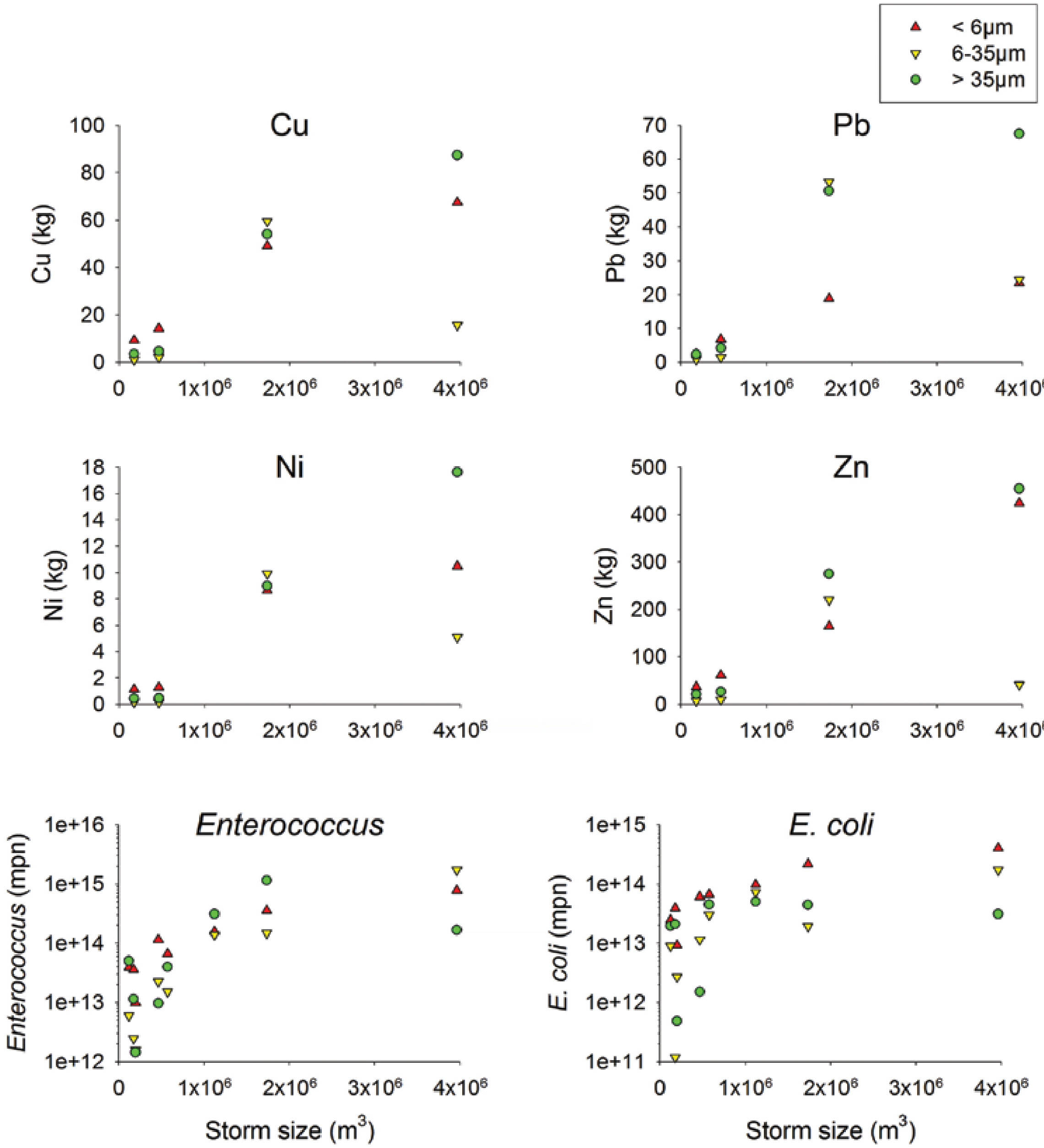

Figure 5. Association of metals and bacteria with particle size relative to storm size.

Ni and Zn were dissolved, but that Pb was equally split between the dissolved and particle-bound fractions. We found a similar trend, with Cu, Ni and Zn associated with the <6 µm filter fraction and Pb concentrations divided between the <6 µm and >35 µm filter sizes in stormwater from Ballona Creek. Likewise, the majority of bacteria were associated with the <6 µm filter fraction, but it is unclear if the bacteria were bound to the smallest particle sizes, or free floating. Krometis *et al*. (2007) determined that approximately 60% of enterococci and *E. coli* were associated with the "suspended" fraction of runoff from low-density residential and institutional (university) drainage areas. Jeng *et al*. (2005) determined that 91% of enterococci and 78% of *E. coli* were not associated with particles in stormwater. Of those organisms that were attached to particles, most of the enterococci were associated with particles between 10 - 30 µm, while *E. coli* tended to attach to particles over a wider range in size (0.45 – 30 µm). Therefore, it is possible that bacteria in the <6 µm fraction in Ballona Creek may have been freely suspended, rather than particle-bound, and may have been associated with dislodged biofilms coating wetted areas of storm drain pipes as suggested by Skinner *et al.* (2010).

In order to evaluate the potential for toxicity in the receiving water, the total metal EMC values were compared to California Ocean Plan thresholds (SWRCB, 2005). Marine receiving water standards were used for the evaluations instead of the freshwater standards because Ballona Creek is not expected to have much water contact recreation compared with the nearby coastal environment, especially during storm events. Daily maximum thresholds were used for comparison: Cu = 12 µg/L, Pb = 8 µg/L, Ni = 20 µg/L, Zn = 80 µg/L. Each of the storms had metal concentrations that exceeded toxicity threshold values. All of the Cu, Pb and Zn EMCs exceeded the daily maximum limits. Cu and Pb EMC values exceeded these thresholds by up to a factor of eight, while Zn EMCs exceeded by up to a factor of five. Ni EMC values did not exceed the threshold in any of the storms sampled.

The bacteria values were compared to water quality standards established to protect water contact recreation in coastal waters (SWRCB 2005). The EMCs for total enterococci were compared to the enterococci 30-day mean standard (35 mpn/100 ml), while total *E. coli* was compared with the fecal coliform 30-day mean standard (200 mpn/100 ml). Objectives do not currently exist for *E. coli* in the marine environment, however because *E. coli* makes up a proportion of the total fecal coliforms, the fecal coliform objective would be a conservative threshold for comparison. Bacteria concentrations exceeded the water quality objectives during each of the storm events sampled. Enterococci EMCs exceeded water contact objectives by up to a factor of 2,750. For *E. coli*, EMCs exceeded the water contact objective by up to a factor of 179.

There has been increasing desire to remove contaminants in runoff using BMPs before the stormwater reaches receiving water. Structural BMPs (including hydrodynamic settling chambers) have been installed at inlets to Ballona Creek in order to capture particulates, trash, and debris (Brown and Bay 2005). However, most of these devices are only able to capture the larger sand particles (≥250 µm), without removing contaminants that are either dissolved or bound to small particulates (Smith 2002). In this study, more than 50% of the Cu, Ni, Zn, *Enterococcus* and *E. coli* were associated with filter size fractions that would not have been captured by these devises. Studies of metals accumulation in urban soils have shown that trace metals primarily accumulate in clay, fine silt, and very fine sand fractions prior to washoff or resuspension as dust (Luo *et al*. 2011, Lau and Stenstrom 2005). Alternative structural BMPs, including media filters, have been developed to capture dissolved contaminants. Using these devices during the initial portions of storms would maximize contaminant removal efficiency, since the highest concentrations of metals in this study were found in the early part of the hydrograph. Storm size should also be considered since metals and bacteria tended to be associated with larger particle sizes during the larger storm events.

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