Particle Size Distribution in Highway Runoff

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Abstract: Particles in highway runoff contain various sorbed pollutants, and many best management practices (BMPs) are selected for particle removal efficiency, which makes particle size distribution a crucial BMP design parameter. Particles between 2 and 1,000 μm in diameter were quantified for three rainfall events during the 2002–2003 rainy season at three highway sites in west Los Angeles. Rainfall, runoff flow rate, and a large suite of water quality parameters were also measured. An experimental protocol was developed for bottle cleaning, sample storage, and mixing that provided repeatable results. Particle aggregation occurred which required samples to be analyzed in less than 6 h; the concentration of small particles decreased with a corresponding increase in the concentration of larger particles in stored samples. The particle concentration decreased as the storm progressed and the number of large particles decreased more rapidly than the total number of particles. Particles demonstrated a strong first flush. On average, 40% of the particles were discharged in the first 20% of the runoff volume.

DOI: 10.1061/(ASCE)0733-9372(2005)131:9(1267)

CE Database subject headings: Stormwater management; Highways; Runoff; Particle size distribution; Best management practice; Pollutants.

Introduction

A great proportion of pollutants in highway runoff such as heavy metals and polynuclear aromatic hydrocarbons are bound to particles (Oliver et al. 1974; Herrmann 1981; Ongley et al. 1981; Hoffman et al. 1985; Hewitt and Rashed 1992; Legret and Pagotto 1999). Most researchers characterize particles as suspended solids (Gupta and Saul 1996; Uchimura et al. 1997; Furumai et al. 2002). The large surface-to-volume ratios of particles in highway runoff provide reactive locations for partitioning and transport of pollutants, and may serve as reservoirs of these pollutants in downstream locations (Oliver et al. 1974; Thomson et al. 1997; Cristina et al. 2002). In addition, pollutants sorbed to particles generally have less mobility and bioavailability than in their dissolved form. Consequently, understanding characteristics of particles in highway runoff is crucial for future runoff management and best management practice (BMP) selection.

Particles in highway runoff arise from roadway maintenance operations, atmospheric deposition, corrosion and erosion, and various kinds of traffic activities such as tire abrasion, vehicular wear, fluid leakage, and pavement degradation (Kobriger and Geinopolos 1984; Thomson et al. 1997; Legret and Pagotto 1999; Grant et al. 2003). Tire and pavement wear produces numerous particles with diameters from several nanometers to several millimeters. Their properties range from quickly dissolving to insoluble (Sansalone and Buchberger 1997b).

Tire and pavement abrasion is the source of many of the particles (Muschack 1990; Sansalone and Tribouillard 1999). Kobriger and Geinopolos (1984) reported the distribution of particles from vehicle-related deposition processes: 37% arise from pavement wear; 37% from tire wear, and 18.5% from abrasion of vehicle parts, such as brakes and engines. Deposition from settleable exhaust accounts for 7.5% of the total particulate mass.

The focus of this study is to characterize particles size distribution (PSD) of highway runoff with major emphasis on developing a protocol to ensure proper particle analysis and reproducible results. In addition, runoff samples were collected throughout the hydrographic and particles were analyzed to verify the existence of particle first flush.

Review of Particle-Sizing Technique

Several particle-sizing techniques have been utilized to characterize stormwater. Sieving techniques use screens for analyzing dry or wet particles. Sedimentation methods have been used for particles in the water column (Gromaire-Mertz et al. 1999). A large variety of instruments have been developed for characterizing particles in the water column, and their advantages and disadvantages are shown in Table 1.

Sieving methods are used for larger particles (generally larger than 45 μm). Ellis and Revitt (1982) used oven drying and sieve analysis for sediments from roadway runoff. Lau and Stenstrom (2001) reported PSD of samples collected during dry weather from roads; they used air drying and mechanical screens over a range of 43–2,200 μm. Sansalone and Buchberger (1997a) collected sediments from individual storm events and measured

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Note. Discussion open until February 1, 2006. Separate discussions must be submitted for individual papers. To extend the closing date by one month, a written request must be filed with the ASCE Managing Editor. The manuscript for this paper was submitted for review and possible publication on August 14, 2003; approved on February 3, 2004. This paper is part of the ***Journal of Environmental Engineering***, Vol. 131, No. 9, September 1, 2005. ©ASCE, ISSN 0733-9372/2005/9-1267–1276/ $25.00.

Table 1. Particle Sizing Techniques Commonly Used in Stormwater Area (Grant et al. 2003)

Particle's (*P*'s) property measured	Aspects measured	Advantages	Limitations	Sample instruments
Transport property: sedimentation	Gravity	Directly applicable results to sedimentation basin design.	Slow	MICROMERITICS Sedigraph
Electrical property: differential resistance	Voltage pulse (proportional to *P*'s volume)	Change of particles in subsize region has no effect elsewhere. Results are not affected by *P*'s shape, nature, gravity, and refractive index.	Carrier fluid influence (e.g., coagulation); May disrupt fragile flocs.	COULTER Multisizer 2
Light obscuration (blockage)	Voltage pulse (proportional to *P*'s maximum cross-sectional area)	Change of particles in subsize region has no effect elsewhere. Results are not affected by *P*'s, nature, gravity, and refractive index. Optical analogue of resistive pulse technique but without electrolyte.	May disrupt fragile flocs.	NICOMP AccuSizer780 PACIFIC SCIENTIFIC INSTRUMENTS Model 9703
Light diffraction property:light intensity	Light intensity	Do not require calibration step.	Concentration of solution has great influence on results.	SEQUOIA LISST-100 MASTERSIZER S Laser Particle Size Analyzer
Dynamic light scattering property: Time or spatial fluctuations in scattering intensity	Hydrodynamic effect—photopulse signal	Good for small particles till 1 nm.	Need long time to get stable	NICOMP PSS 170

PSD by drying the sediments at 110°C and sieving with mechanical screens. The drying step required before sieving can alter particle size and character (Krein and Schorer 2000). In addition, due to potential aggregation among particles, PSD in sediments does not necessarily represent the PSD in runoff (Slattery and Burt 1997).

Roberts et al. (1988) utilized a Coulter Counter with electrical resistance technique to measure particle sizes and employed scanning electron microscopy to characterize the alteration of runoff-entrained pavement solids transported through a pipe sewer during rainfall runoff events. Sansalone et al. (1998) investigated PSD in highway runoff using a HIAC/ROYCO light obscuration instrument and mechanical sieves. Drapper et al. (2000) employed a Mastersizer laser particle size analyzer to determine the PSD of roadway runoff samples taken from sites in southeast Queensland, Australia. The instrument uses light diffraction technique.

Hargesheimer et al. (1992) proposed an experimental particle-sizing protocol for enumerating the total number of particles in wastewater effluents, and described sample collection, handling and analysis, and storage procedures. They found that although carefully cleaned caps and freshly dispensed parafilm contaminated samples with small particles (most less than 1 μm in diameter), glass bottles, cleaned by hand, dishwasher, or supercleaning procedures, were satisfactory sample containers when counting particles as small as 0.5 μm in diameter. Plastic and teflon bottles were unsuitable containers when counting particles less than 5 μm in diameter, regardless of cleaning methods. Gentle inversion, sonication, and inversion–sonication–inversion were satisfactory mixing techniques, producing similar particle counting results. Gentle inversion was preferred because of its simplicity. They found that the variability of particle counts increased with storage time and results were most reproducible immediately after sample collection. Although several researchers have measured PSD in highway runoff or stormwater (Characklis and Wiesner 1997; Sansalone Buchberger 1997a; Sansalone et al. 1998; Legret and Pagotto 1999), no consistent experimental method has evolved. In addition, few researchers have counted PSD over a wide range (2–1,000 μm) and over entire storm events. Both of these topics are the focus of this paper.

Methodology

Site Description

Three monitoring sites in west Los Angeles were selected with catchment areas ranging from 0.39 to 1.69 ha and annual average daily traffic of over 260,000 vehicles/day. These three sites were chosen as typical small catchment area sites with heavy traffic load, and were within 15 min travel time to the lab at Univ. of California, Los Angeles. The sites were so small that there was very little delay between the peak rainfall and peak runoff (i.e., 5 min or less). All sites were equipped with American Sigma (Loveland, Colo.) 950 flow meters, tipping bucket rain gages, and composite auto samplers. For additional information on composite sampling equipment, the reader can refer to Kayhanian et al. (2003). Additional site descriptions are summarized in Table 2.

Sample Collection Procedure

Grab samples were collected manually with a polypropylene container from a free waterfall as runoff exited the drainage pipe, and stored in 4 L narrow mouthed amber glass bottles. Collection began immediately after the beginning of runoff, usually within a few minutes of the beginning of rainfall. Subsequent samples were taken during the first hour at 15 min intervals. After the first hour, grab samples were taken at 1 h intervals for the following 7 h. Some storm event durations in the study area were more than 8 h. For storms lasting longer than 8 h, one or two additional grab samples were collected. Flow-weighted composite samples were collected using several 4 L glass bottles by composite auto

Table 2. Site Description Summary

Site ID	Location	AADT (vehicles/day)	Catchment area (ha)	Number of lanes (/direction)	Drainage pipe diameter (mm)	Approximate impervious (%)
1	Hwy 101, Van Nuys, Calif.	328,000	1.28	6	508	100
2	Hwy 405, Getty Center Exit	260,000	1.69	5	610	95
3	Hwy 405, Santa Monica Blvd.	322,000	0.39	5	600	100

Note: ID=identification and AADT=annual average daily traffic.

samplers. The grab samples collected within the first hour were delivered to the lab (15 min or less travel time) once the fifth grab sample was taken. The PSD was analyzed as soon as the samples reached the lab and completed within the next 2 h. The samples collected from the second hour until the end of the storm event were periodically taken to the lab, and all were analyzed for PSD within 6 h of collection. Composite samples were also brought to the lab at the end of storm event and were analyzed within 6 h of collection. The logic behind the PSD analysis within 6 h is discussed in the following section.

Particle Size Analysis

A Nicomp Particle Sizing Systems (Santa Barbara, Calif.) AccuSizer 780 optical particle sizer module equipped with an autodilution system and a light scattering/extinction sensor (Model: LE1000-2SE) was used for particle size analysis. This instrument was selected for its wide range (2–1,000 μm), speed (<2 min/sample analysis), and auto dilution capability. A representative sample ranging in volume from 1 to 10 mL was removed from the 4 L sample bottle using a wide-bore glass pipette, after gently inverting the 4 L bottle 5–6 times, and then injected into the AccuSizer. Between samples, the system was flushed at least three cycles, which reduced background particle concentrations to less than 3/mL.

Results and Discussion

Particle Size Distribution Protocol Development

As discussed in the previous section, no standard protocol exists for measuring PSD in stormwater. A series of experiments was performed to establish a standard protocol with known accuracy and repeatable results. These experiments were performed to understand four key sampling concerns: PSD reproducibility, sample contamination, sample representativeness, and sample storage time and temperature. These parameters were studied for 3 months using stored samples, before the start of similar experiments with fresh highway runoff samples. Each experimental parameter is discussed in the following sections.

Particle Size Distribution Reproducibility

The reproducibility of the PSD obtained by AccuSizer was demonstrated by measuring duplicate stormwater samples. Fresh samples were randomly obtained from three storm events. The difference between each duplicate pair was represented by a difference proportion (DP), calculated as follows:

$$\text{difference proportion} = 100\frac{2|N_1 - N_2|}{(N_1 + N_2)} \quad (1)$$

where N_1 and N_2=number of particles in a specific size range for the first and second samples. The mean and variance of DP values of 11 duplicates are shown in Table 3. The difference for duplicate samples was within 10% for particles less than 30 μm. The DP increased for larger particles, and the difference was approximately 76% for particles in the range of 200–1,000 μm. This resulted in part because there were less particles in this size range. The number of particles with diameter from 200 to 1,000 μm ranged from 0 to 57/mL with mean value 8/mL. This means that even a small difference in particle number will produce a large DP. For the 2–3 μm size range, the number of particles averaged 139,000/mL. To decrease the variability of the large particle measurements, a greater number of samples should be collected or the measuring range should be modified to include more particles. We did not modify our procedure to reduce the variability of larger particle measurement since the smaller particles (<200 μm) were most abundant and are the focus of our study and many other recent projects (Furumai et al. 2002; German and Svensson 2002; Sutherland 2003). Additionally, the larger particles have lower pollutant concentrations (Sansalone and Buchberger 1997a; Roger et al. 1998; Lau and Stenstrom 2001; German and Svensson 2002; Morquecho and Pitt 2003).

Sample Contamination

We investigated four possible sources of particle contamination. These four sources included: a grab sampling device (i.e., scoop 160 mm×120 mm×200 mm), sample bottle (4 L glass bottle and cap), transfer pipette, and instrument (dilution chamber and stirrer). The sampling scoop was eliminated as a source of contamination by rinsing the scoop in fresh runoff prior to sample collection. The particle-sizing instrument was eliminated as a

Table 3. Mean and Variance of Difference Proportion (DP) Values and Mean Particle Number for 11 Duplicate Samples

Statistical parameter	Particle diameter range (μm)										Total
	2–3	3–5	5–7	7–10	10–20	20–30	30–50	50–100	100–200	200–1,000	2–1,000
DP mean	9.5	4.5	6.1	8.3	9.6	9.7	20.4	35.4	66.9	75.6	5.0
DP variance	0.5	0.1	0.2	0.5	0.8	0.9	2.4	12.0	24.4	64.2	0.1
N mean (number/mL)	138,725	119,219	38,507	20,233	12,858	1,561	363	57	19	8	331,551

Table 4. Particle Number Concentration Based on Contamination Experiment (Number/mL)

Sample		Particle diameter range (μm)										Total
		2–3	3–5	5–7	7–10	10–20	20–30	30–50	50–100	100–200	200–1,000	2–1,000
Instrument[a]	I1	2	0	0	0	0	0	0	0	0	0	2
	I2	1	0	0	0	0	0	0	0	0	0	1
Beaker	B1	12	10	6	5	6	2	3	1	2	2	49
	B2	12	8	6	3	3	1	2	1	1	1	38
	B3	13	8	3	3	3	2	4	1	1	1	39
4 L bottle	Bo1	14	10	9	5	8	3	4	0	1	1	55
	Bo2	45	33	16	8	5	2	2	0	0	1	112
	Bo3	81	75	34	20	12	2	1	1	3	2	231
	Bo4	27	29	16	13	11	4	3	0	0	1	104

[a]Without sample injection.

source by flushing the chamber through at least three cycles with de-ionized (DI) water filtered through a 0.2 μm membrane filter, which usually reduced particle counts to less than 3/mL (Table 4). Contamination from sample bottles and glassware was evaluated by counting particles in cleaned bottles and beakers. Sample bottles were cleaned by soaking overnight in detergent, rinsing with hot water 5–6 times, rinsing with 10% nitric acid, rinsing with dichloro-methane, rinsing in DI water, and oven drying. Four bottles (4 L) and three 200 mL beakers were evaluated. The beakers were cleaned by soaking overnight in detergent, rinsing with hot water 5–6 times, and rinsing with nanopure, particle free water (NPPFW) obtained from a Barnstead Nanopure Infinity Water System with a 0.2 μm filter. All bottles and beakers were filled with NPPFW.

Each container was sampled with a 10 mL pipette and analyzed. The results are shown in Table 4. After cleaning, the instrument was essentially particle free, which was treated as background concentration. The beakers had very few particles, ranging from 38 to 49/mL. The bottles had particle concentrations ranging from 55 to 231/mL. This procedure evaluated the combined contamination sources of cleaning, bottle, cap, and pipette.

Particle concentration decreased with increasing particle diameter. Instrument particle concentration was 2/mL or less, which was satisfactory. The washing procedures cleaned the bottles to less than 250/mL total particles in all size fractions. The cleaning method was adopted since runoff particle concentrations were usually in excess of 10,000/mL.

Sample Representativeness

Sample representativeness is very important to characterize highway runoff PSD because particle-sizing instruments typically need only a small amount of sample injection. In addition, runoff from highways usually contains particles that will settle in a 4 L bottle within 10–20 s or less. Complete mixing of samples is therefore an essential step to assure a representative sample. Four procedures were used to mix the 4 L sample bottles: (1) no mixing; (2) inversion by gently inverting the 4 L bottle 5–6 times in 30 s; (3) gentle inversion–decanting–stirring by using gentle inversion as in (2), following by removing 100 mL of the sample into a beaker on a magnetic mixer and agitating for 10–20 s at 400 rpm; and (4) gentle inversion–decanting–blending, similar to (3), except that 200 mL of sample was poured into a clean blender (Waring commercial blender) and blended at 3,500 rpm for 1 min. In each procedure, the end of a large-bore pipette was inserted 2 cm below the liquid surface to collect a 1–10 mL sample immediately after the final mixing step, and injected into the particle sizing system for measurement. Fig. 1 shows the PSD of the above four analytical preparation techniques. Without inverting the sample bottle, the particle measured concentration was far less than measured with mixing, suggesting sedimentation in the bottle. Gentle inversion and gentle inversion–decanting–stirring produced similar PSD results. Blending destroyed many large particles, producing many additional small particles (Laubscher et al. 2001).

To further evaluate gentle inversion and gentle inversion–decanting–stirring, PSDs of four samples were analyzed using both methods, and the results are shown in Table 5. Concentrations of particles with diameters between 2 and 1,000 μm measured with the two methods produced DP values less than 3. The large DP values of particles with diameters greater than 100 μm are due in part to the low concentration of larger particles, as described before in the reproducibility analysis. The gentle inversion and gentle inversion–decanting–stirring methods produced identical results and gentle inversion was adopted for this study.

Sample Storage Time and Temperature

The PSD may change with storage time because of particle aggregation or dissolution. To quantify the changes in PSD with storage time, seven grab samples taken on November 7, 2002 and

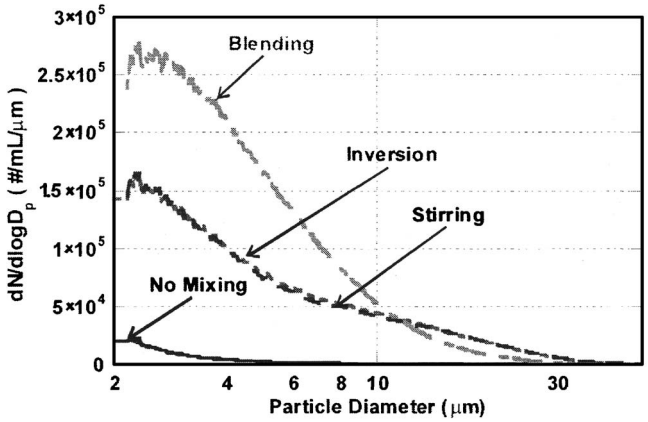

Fig. 1. Comparison of different mixing methods (labels describe various methods—see the text for their description; dN=number of particles per milliliter within certain size range D_1–D_2, $d \log D_p = \log D_2 - \log D_1$)

Table 5. Concentration Difference Evaluation Using Inversion and Stirring Methods

Diameter range (μm)	Sample 1			Sample 2			Sample 3			Sample 4		
	Inversion[a]	Stirring[a]	DP	Inversion[a]	Stirring[a]	DP	Inversion[a]	Stirring[a]	DP	Inversion[a]	Stirring[a]	DP
2–1,000	176,601	181,548	2.8	611,184	603,383	1.3	94,741	95,276	0.6	96,090	95,533	0.6
2–3	42,294	44,351	4.7	309,057	307,472	0.5	39,423	39,959	1.4	42,813	41,785	2.4
3–5	57,797	60,073	3.9	222,907	220,401	1.1	34,558	34,921	1.0	36,709	36,435	0.7
5–7	31,622	32,492	2.7	50,004	48,569	2.9	10,865	10,902	0.3	10,518	10,845	3.1
7–10	23,712	24,054	1.4	19,234	17,744	8.1	5,557	5,418	2.5	4,167	4,507	7.8
10–20	17,698	17,316	2.2	8,461	7,918	6.6	3,497	3,304	5.7	1,749	1,829	4.5
20–30	2,407	2,213	8.4	983	782	22.8	594	499	17.4	97	99	2.0
30–50	827	802	3.1	341	338	0.9	215	225	4.5	31	27	13.8
50–100	207	209	1.0	120	115	4.3	29	42	36.6	4	4	0.0
100–200	31	34	9.2	48	42	13.3	3	5	50.0	1	2	66.7
200–1,000	6	4	40.0	29	2	174.2	0	1	200.0	1	0	200.0

Note: DP=difference proportion.

[a]Unit=number/mL.

December 15, 2002 were kept in a 4°C cooling room. At the same time, 200 mL of sample were poured into beakers from each of the seven grab samples after gentle inversion, and kept at room temperature (20°C), open to the atmosphere, but protected from dust fall or any other contamination. Fig. 2 shows the PSD over time for one sample, stored at 4 and 20°C. It can be seen that the particle size generally increased over time at both storage temperatures. The size of the particles in the sample stored at room temperature increased at a much higher rate. Particle size measurements were terminated after 60 h under room temperature due to particle breakup during analysis.

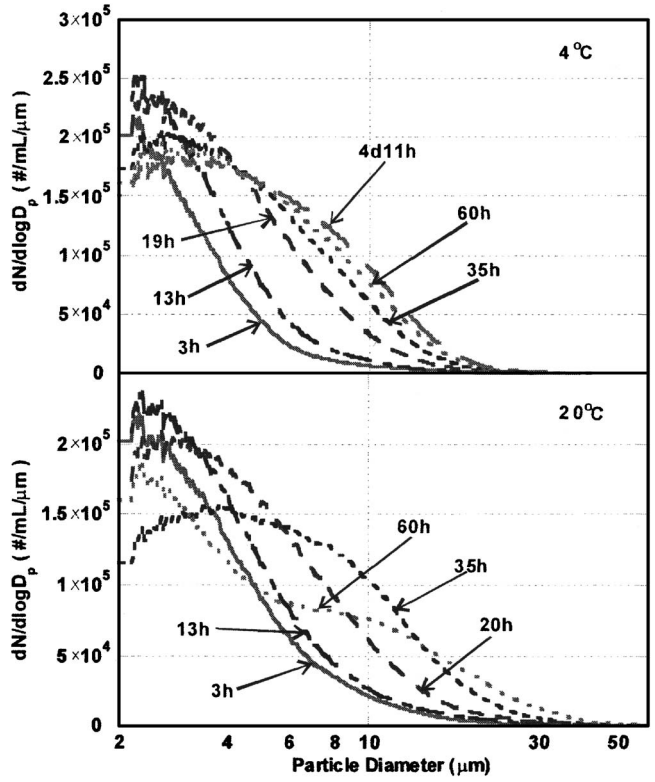

Fig. 2. Sample storage time and temperature influence (Site 3, event December 15, 2002, grab sample number 2, labels show storage time in hours)

To illustrate the changes in particle sizes, the numbers of particles in specific size fractions at different times were normalized by dividing by the initial particle concentration. The normalized concentrations of all seven grab samples were averaged and plotted as ratios. Fig. 3 shows the refrigerated samples. Concentrations of particles in the smallest fractions increased for the first 50 h and then decreased. The particles stored at room temperature (Fig. 4) showed the same trend but at a much more rapid rate with larger increases in concentration. Particles larger than 7 μm at both temperatures showed a monotone increase in particle numbers through the period of observation.

The rapid growth in particle size suggests a naturally occurring coagulation/flocculation mechanism. The presence of naturally occurring flocculation and an increase in particle size can have a

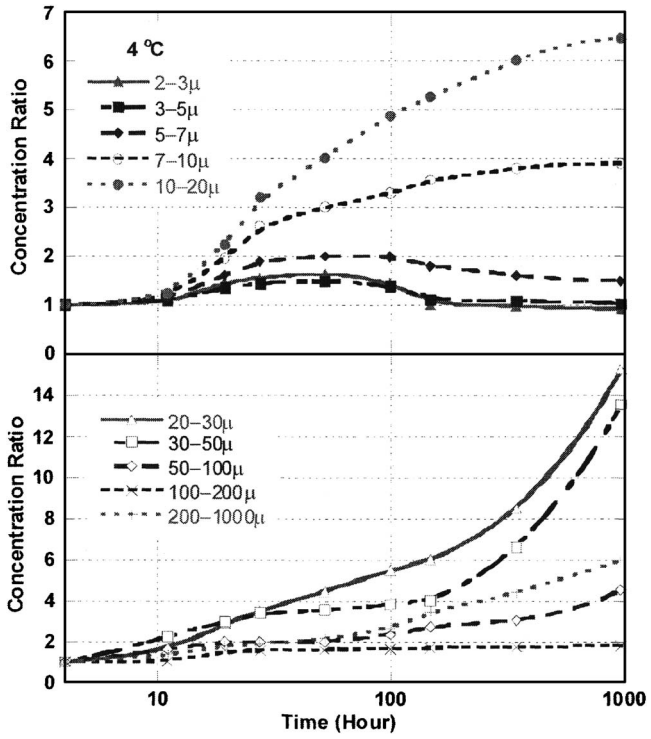

Fig. 3. Average concentration ratio versus time (4°C)

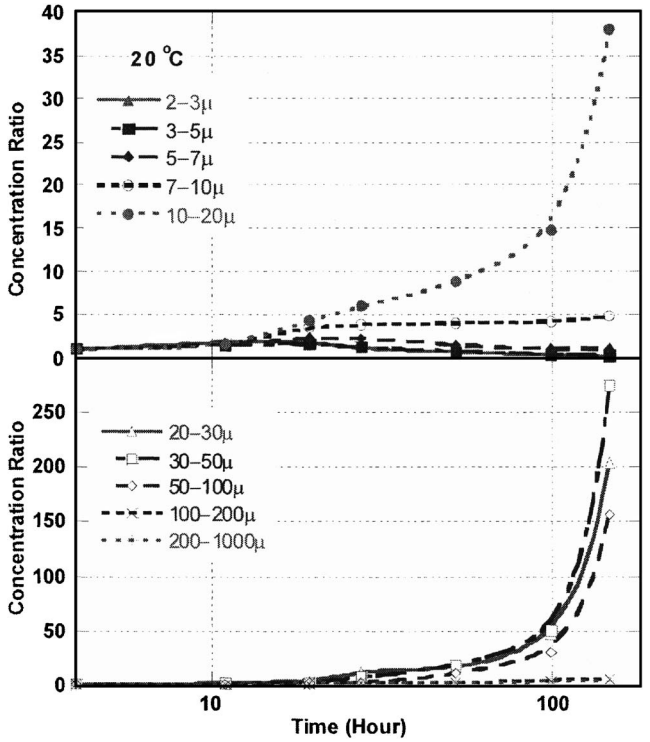

Fig. 4. Average concentration ratio versus time (20°C)

profound impact on sample storage and hence on designing stormwater treatment systems (e.g., BMPs). Based on these results, all samples for PSD analysis were analyzed within 6 h of collection. Treatment systems that hold the stormwater for appreciable amounts of time may have improved sedimentation rates due to particle growth.

The number of particles in the range of 2–7 μm increased in all samples at the beginning of storage. Particle numbers continued to increase for 13–50 h, depending on the sample. After this time, the numbers gradually decreased. This suggests that new particles were being formed from precipitation, or that particles too small to be counted were increasing in size and appearing in the smallest fraction. Future work should investigate this phenomena, since particles in this range will likely escape most treatment devices except for those that provide soil infiltration. Accelerating particle aggregation should help overall BMP effectiveness for the removal of solids and pollutant load that are associated with those solids.

Particle Size Distribution with Respect to Hydrograph and Partial Water Quality Parameters

Table 6 summarizes three events where PSD was monitored. Fourteen grab samples were collected at all three sites on November 7, 2002. The other two storms were shorter and fewer samples were collected. Turbidity, conductivity, and total suspended solids (TSS) were measured and reported in this paper. A large suite of other parameters were measured and reported elsewhere (Ma et al. 2002).

Fig. 5 shows the PSD, rainfall, runoff, TSS, turbidity, and conductivity for Site 1, event November 7, 2002 as a representative example of the three events. The concentration of particles (2–1,000 μm) in 61 grab samples of all three events ranged between 50,000 and 3,742,000/mL with a median value 204,000/mL. For all events, the highest particle concentration always occurred within the first hour, and decreased rapidly thereafter.

The lower two graphs in Fig. 5 show PSD for the composite sample and various grab samples. The data are plotted on two axes for clarity. Each distribution has a time label that can be compared to the top of Fig. 5 to show the point on the hydrograph when the sample was taken. The trends shown are typical of most samples (three out of six storms). The very first grab sample did not have the highest particle concentration (number of particles per base-10 logarithm of particle diameter per milliliter), which probably resulted because runoff had not developed this early in the storm. Particle concentration increased to a maximum over the next 15–45 min, and generally decreased in later samples. The vast majority of the particles were less than 10 μm, and after several hours particles larger than 10 μm were reduced in concentration. The median particle size decreased as the storm progressed, which shows a more rapid washout of larger particles.

In many of our previously sampled storms, we observed increases in contaminant concentrations after rapid increases or "spikes" in runoff flow. At approximately 13 h such a spike occurred for example (Fig. 5), which increased particle concentration and turbidity. An examination of many data sets does not reveal a strong correlation between instantaneous runoff flow rate and pollutant concentrations, but there are many examples of spikes in flow rate followed by spikes in concentration (Uchimura et al. 1997), particularly suspended solids. It is believed that there is a cause and effect relationship between instantaneous runoff rate and contaminant concentration, but a quantitative relationship has not yet been found.

Conductivity decreased rapidly as the storm progressed, and is typical of many of our observations; conductivity usually exhibits a significant first flush, which was evaluated with the mass first

Table 6. Event Summary

Monitoring site	Event date	Grab sample number	Antecedent dry day (days)	Event rainfall (mm)	Runoff volume (m^3)	Maximum intensity (mm/h)	Rainfall duration (h:min)	Runoff duration (h:min)
1	November 7, 2002	14	40.1	29.0	210.5	10.2	47:31	44:09
2	November 7, 2002	14	41.2	58.7	825.8	14.2	46:29	46:43
3	November 7, 2002	14	40.2	71.4	178.0	12.2	47:05	47:38
2	November 29, 2002	5	20.2	1.8	23.3	3.1	7:44	8:14
3	November 29, 2002	6	20.2	1.5	0.7	2.0	6:52	7:21
2	December 15, 2002	8	16.1	2.5	30.3	2.0	3:20	4:38

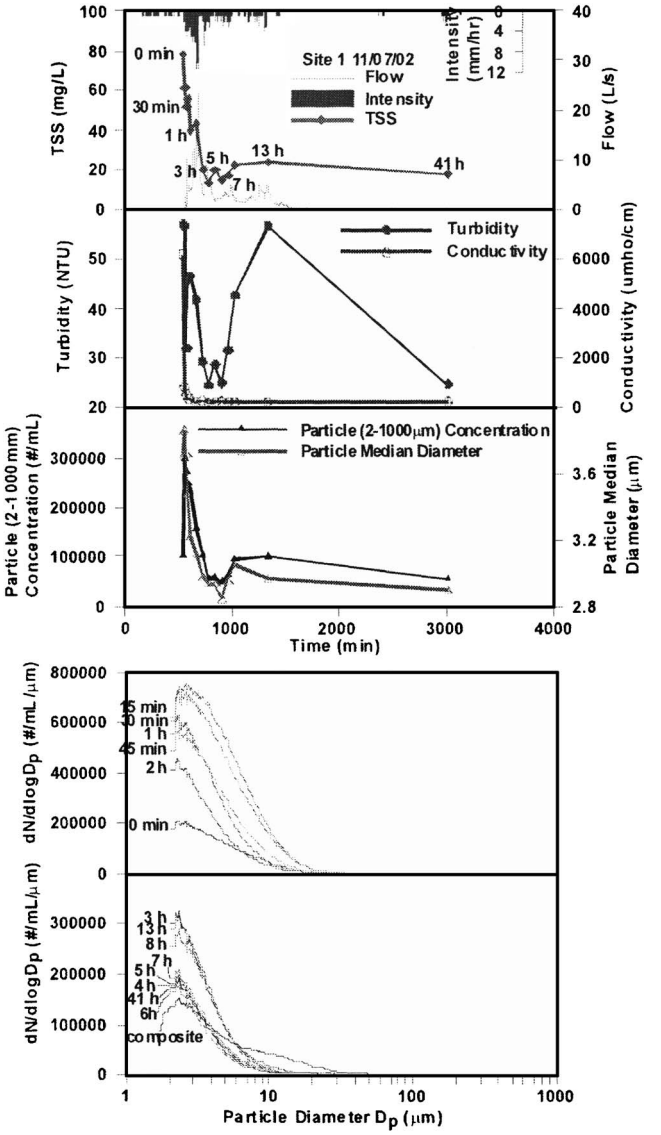

Fig. 5. Hydrograph with total suspended solids, turbidity, conductivity, and particle size distribution for Site 1, event November 7, 2002

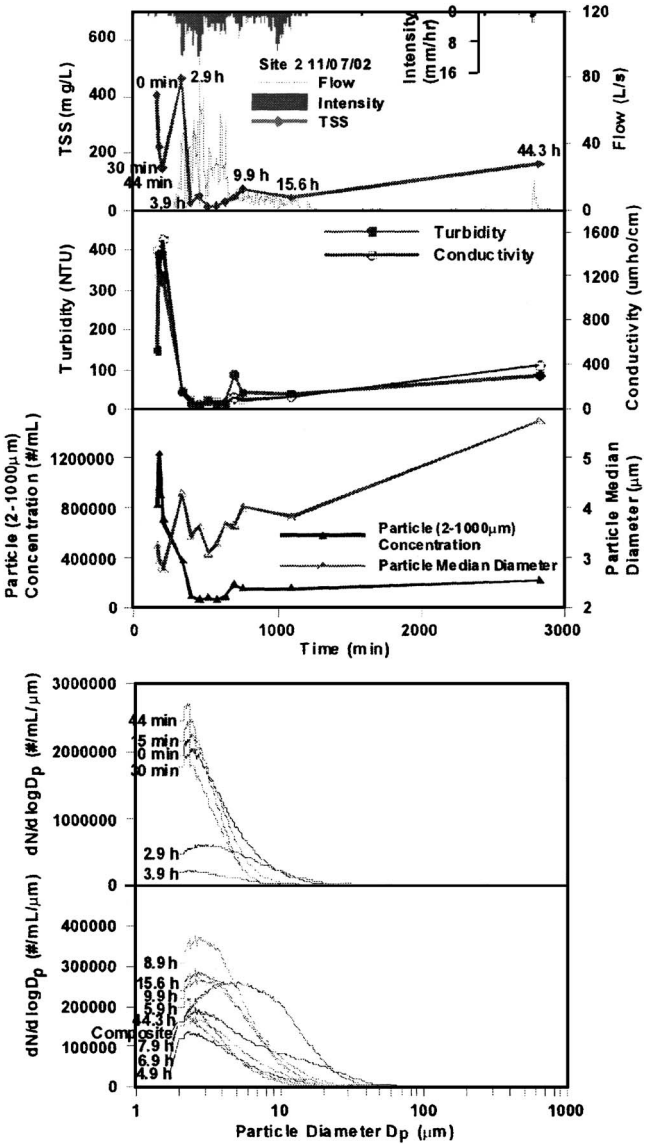

Fig. 6. Hydrograph with total suspended solids, turbidity, conductivity, and particle size distribution for Site 2, event November 7, 2002

flush (MFF) ratio developed by Ma et al. (2002). The MFF ratio was defined as the normalized mass fraction divided by the normalized volume fraction at any given point along the normalized or fractional runoff diagram. It is thought that the low conductivity rain water eventually dilutes out the salts being flushed from surfaces.

Fig. 6 shows similar results for Site 2, event November 7, 2002. Early samples had the greatest particle concentration. Median particle diameter did not decline but increased slightly. The increase at the end of the storm is due to the brief, intense rainfall at 44 h. The rainfall is responsible for increases in TSS, turbidity, and particle concentration. Fig. 7 shows the results from Site 3 on November 29, 2002. This was a shorter storm with reduced rainfall. It is easy to observe the washout of small particles as the storm progressed. There is a surge in TSS, particle concentration, and median diameter at 6.75 h, which corresponds to additional rainfall.

The particle concentrations in composite samples are also shown in Figs. 5, 6, and 7. They are lower than most grab samples, and should have a particle concentration between the lowest and highest. The composite sample concentrations were only between 34 and 94% of the corresponding flow-weighted averages of the grab samples. This large difference suggests that aggregation or some other phenomena is occurring. Composite samplers may not be the most appropriate method of collecting samples for PSD analysis due to potential aggregation between particles.

These PSDs of grab sample results can be compared to other results if particle number concentrations are converted to mass concentration by assuming spherical particles and uniform particle specific gravity for particles with diameters between 2 and 1,000 μm. Lau and Stenstrom (2001) recovered particles from dry pavement, and found that only 3% of the particle mass occurred in particles smaller than 50 μm. Sansalone and Tribouillard (1999) observed less than 10% of total particle mass in particles smaller than 50 μm. This research suggests that nearly 30–60% of the particle mass is found in particles smaller than 50 μm (Fig. 8).

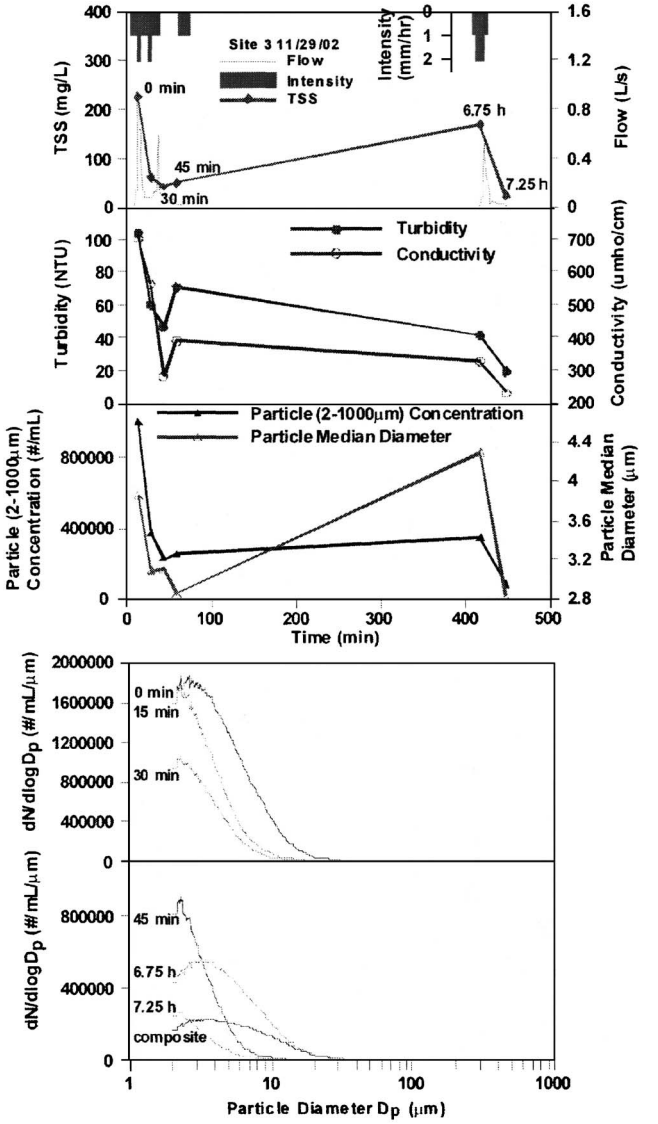

Fig. 7. Hydrograph with total suspended solids, turbidity, conductivity, and particle size distribution for Site 3, event November 29, 2002

The removal of particles by BMPs is highly dependent on the particle size, shape, and specific gravity. It is also important to know pollutant distribution among the various particle size fractions. The combination will allow BMPs to be selected to optimize pollutant removal as well as particle removal. This topic is currently under investigation in our laboratory.

First Flush of Particles

First flush phenomenon is a controversial topic in stormwater management (Characklis and Wiesner 1997; Deletic 1998). Certain researchers (Barrett et al. 1998) found that the overall first flush effect of highway stormwater runoff was small or negligible, while other researchers (Gupta and Saul 1996; Sansalone and Buchberger 1997b; Lau et al. 2002; Ma et al. 2002) observed a moderate or strong first flush. Generally, smaller catchment areas exhibited stronger first flushes (Characklis and Wiesner 1997). In addition, no consensus definition exists for the first flush (Bertrand-Krajewski et al. 1998) and different researchers have used different definitions (Geiger 1984; Thornton and Saul 1987; Ashley et al. 1992; USEPA 1993; Gupta and Saul 1996; Sansalone and Buchberger 1997b, a; Deletic 1998; Ma et al. 2002). In this discussion, we extend the MFF ratio developed by Ma et al. (2002) to the particle first flush, and present it as the particle number first flush (PNFF) ratio.

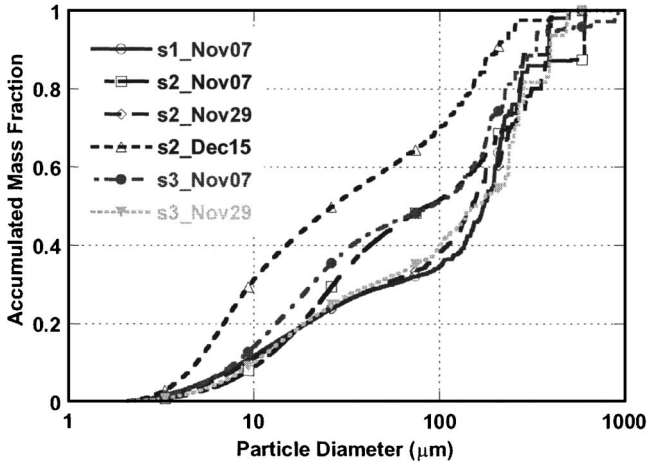

Fig. 8. Accumulated particle size distribution by mass (assuming spherical particles with uniform density through all size ranges; s1_Nov07 indicates event November 7, 2002 of Site 1)

The PNFF ratio is defined as the normalized number of particles divided by normalized volume fraction at any point of the normalized runoff diagram. It is similar to the first flush definition proposed by Bertrand-Krajewski et al. (1998).

Let x represent the x percent runoff volume at a certain time t_1. Then

$$x\% = \frac{\int_0^{t_1} Q(t)dt}{V} \tag{2}$$

$$\text{PNFF}_x = \frac{\dfrac{\int_0^{t_1} C_p(t)Q(t)dt}{N}}{\dfrac{\int_0^{t_1} Q(t)dt}{V}} \tag{3}$$

where $Q(t)$=runoff flow rate (L^3/T); $C_p(t)$=particle number concentration (L^{-3}); V=total runoff volume of an event (L^3); and N=total number of particles in an event.

The ratio can be calculated for any specific point in the storm, and approaches 1.0 by definition at the end of the storm. The ratio allows convenient characterization of the first flush. For example, $\text{PNFF}_{20}=2.5$ means that when the accumulated runoff volume reaches 20% of total runoff volume, it contains $20\% \times 2.5 = 50\%$ of total particle count in an event. The MFF ratio is also defined by Eq. (3), except that concentration is used instead of particle number.

Fig. 9 shows the number first flush ratios ($\text{PNFF}_{10}, \text{PNFF}_{20}, \text{PNFF}_{40}$) for particles in different size ranges. The top and bottom of the box mark the limits of ±25% of the variable population, and the horizontal line is the median. The whiskers represent the maximum and minimum observed values, unless there are outliers. Fig. 9 illustrates that median PNFF_{10},

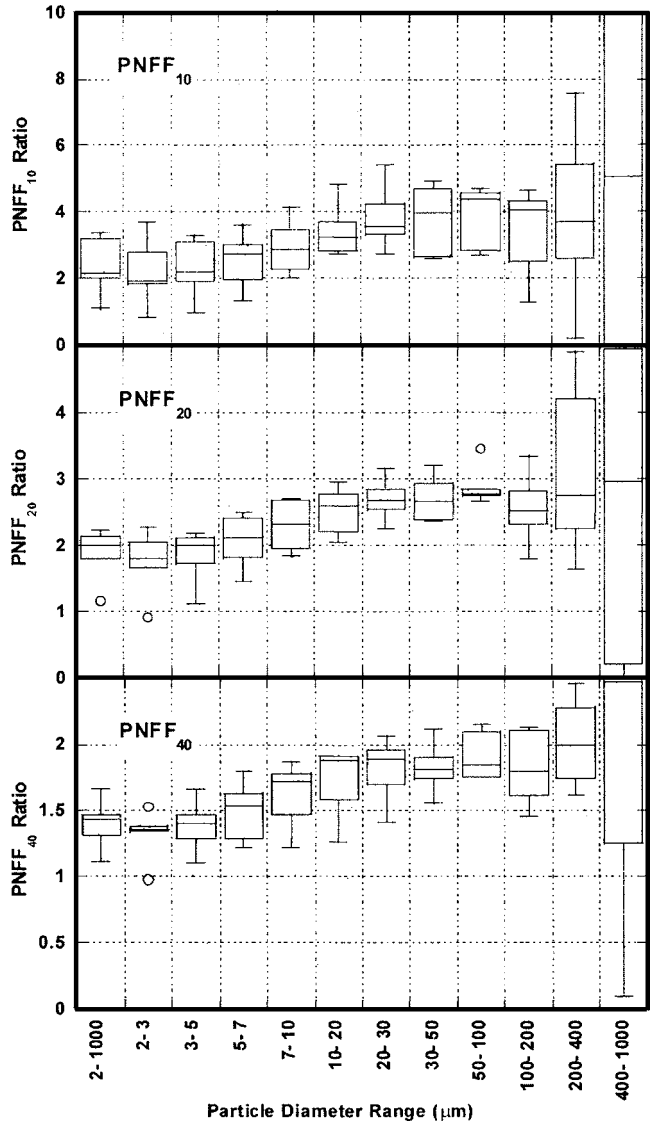

Fig. 9. Boxplots for different particle number first flush ratio

$PNFF_{20}$, and $PNFF_{40}$ values generally increase with increasing particle diameter. The PNFF ratio is generally larger than the analogous MFF ratio for other water quality parameters such as TSS and turbidity (Ma et al. 2002). This suggests that BMPs that can completely capture the early runoff will be more effective than BMPs that treat a portion of the runoff throughout the storm.

Conclusions

The PSD was characterized in three storm events at three sites. It was necessary to develop an experimental protocol to ensure representative samples. A hand washing procedure for glass bottles was a suitable method for preventing contamination of samples being analyzed for PSD; gentle inversion (5–6 times) of the sample bottle was an appropriate mixing method that prevented sedimentation or particle shearing. Particles showed a natural aggregation, which required analysis as soon as possible but within 6 h of sample collection. Particle concentrations in samples collected by the automatic samplers were lower than a flow-weighted average of the grab samples. Results suggest that automatic composite samplers should not be used to collect samples for PSD analysis until further development is completed.

More than 97% of the particles were less than 30 μm. Particle concentration and size generally decreased rapidly as the storm progressed. Rapid increases in particle number occurred after rapid increases in rainfall or runoff, and were accompanied by increases in turbidity and TSS concentration. Particles showed an obvious first flush, with median of $PNFF_{20}$ of approximately 2, indicating that 40% of total particles were carried in the first 20% of runoff volume. Larger particles showed a stronger first flush than smaller particles.

Acknowledgments

This study was supported in part by the California Department of Transportation (Caltrans). The writers are grateful for their continuous support.

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