First flush and natural aggregation of particles in highway runoff

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Abstract Particle Size Distribution (PSD) in highway runoff was monitored in the 2004–2005 rainy season at three highway sites in west Los Angeles, California. PSD was measured for 200 grab samples for 18 storm events. Particles and especially larger particles showed a strong first flush. On average, the initial 20% runoff volume transported approximately 28% total number of particles between 0.5 and 2 μm in diameter, more than 30% of particles between 2 and 30 μm and more than 40% of particles larger than 30 μm. A naturally occurring aggregation was observed with smaller particles and mixing experiments were performed to determine the possible benefits for sedimentation and filtration. Samples composited from grab samples manually collected over the first hour of runoff were gently mixed ($G = 38$) and small particle concentration decreased by more than 50%. After 24 hours the number of particles with diameter between 0.5 and 7 μm decreased by 51% with gentle mixing and the same size particles decreased by only 14% without mixing. Number of particles with diameter larger than 20 μm increased by 6 and 4.5 times with and without mixing, respectively. Slow mixing can improve sedimentation efficiency by more than 40% for particles less than 20 μm in diameter.

Keywords Aggregation; first flush; highway runoff; mixing; particle size distribution; sedimentation

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Introduction

Non-point source pollution has become the major pollution source to the receiving water bodies in certain developed and developing countries. In the United States, urban stormwater runoff contributes to 13% of impaired river and stream miles, 21% of impaired lake areas, 55% of impaired ocean shorelines miles, and 46% of impaired estuary square miles (USEPA, 2000). In the People's Republic of China, non-point source pollution contributes 67% of total nitrogen and 63% of total phosphorus pollution to river bodies (Zhou *et al.*, 2004).

Among various non-point sources, roadway runoff has received much attention due to the presence of heavy metals, which are non-degradable, accumulative and toxic to ecosystems (Barrett *et al.*, 1998). In China, 80% of the sediments in rivers and lakes are polluted by heavy metals (Zhou *et al.*, 2004). For roadway runoff management, fine to medium particle removal has received much attention due to the higher concentration of heavy metals on smaller particles (Sansalone and Buchberger, 1997; Morquecho and Pitt, 2003).

Various stormwater treatment systems have been deployed including wetlands, ponds, biofiltration, vortex or swirl concentrators, detention basins and sand filters (Bäckström, 2003; Lee *et al.*, 2003). Most treatment methods target particle removal and do not impact dissolved pollutants. In general, larger particles are easier to remove than smaller particles. Recent investigation on particle size has shown that particles in highway runoff increase in size with time due to natural aggregation/flocculation (Li *et al.*, 2005).

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This paper documents the natural aggregation and explores the potential benefits for improving the best management practice (BMP) performance.

Methods

Three highway sites (herewith known as sites 1, 2 and 3) in west Los Angeles were chosen as typical small catchment area sites (0.39 to 1.28 hectares) with heavy traffic load (260,000 to 328,000 annual average daily traffic or AADT). At each site, grab samples were manually taken at a free water fall when runoff exited the drainage pipe. The first grab sample was taken at the beginning of runoff, followed by four more grab samples at 15 min intervals over the first hour. Additional grab samples were taken at one hour intervals over the following 7 h. One or two additional grab samples were usually taken after 8 h of runoff for long storms. For additional site description and grab sample collection procedures, please refer to Li *et al.* (2005). A total of 18 storm events were monitored and 200 grab samples were taken from the three monitoring sites during the 2004–2005 rainy season.

PSD was analyzed with a Nicomp Particle Sizing Systems (Santa Barbara, California) AccuSizer 780 optical particle sizer module equipped with an auto dilution system and a light scattering/extinction sensor (Model: LE400-0.5) with measurable range 0.5 to 400 μm. The detailed particle size analysis procedure can be found in Li *et al.* (2005).

To evaluate the effect of natural aggregation, slow mixing experiments were conducted on additional samples manually collected over the first hour from site 2. During the first hour of runoff, one litre additional runoff samples were collected from a free water fall every five minutes after the beginning of runoff. All of the 13 additional grab samples were brought back to the lab immediately and mixed together in a 20 L plastic tank. Two 4 L glass bottles were then filled with this first hour composite sample. A stirring bead was placed into one bottle which was placed on a magnetic mixer. The other bottle was left on the bench without mixing. Particle size distribution (PSD) was periodically analyzed for both samples for 1 to 23 days, depending on the experiment. The G factor was calculated as 38 assuming that the mixing bead could be approximated by a 2 paddle stirrer.

Results and discussion

Particle dynamics

Table 1 describes the characteristics of the monitored storm events. Most of these storm events lasted longer than 8 h. A typical hydrograph and pollutant concentration of grab samples as a function of time are shown in Figure 1. The top of Figure 1 shows the rainfall, runoff, TSS, turbidity, conductivity, particle concentration and particle median diameter of grab samples for Site 2, on October 16, 2004. TSS, conductivity and particle concentration show a strong first flush with the early grab sample(s) having two to eightyseven times higher concentration than later samples. Generally, for most storm events monitored, the highest particle concentration always occurred within the first hour, and decreased rapidly thereafter. The median particle size decreased with the progress of rainfall because the larger particles are discharged early in the storm (i.e. stronger first flush, Li *et al.*, 2005, 2006a).

The lower two graphs in Figure 1 show PSD for each grab sample of the same event. The data are plotted on two *y* axes with different scales for clarity. Each distribution has a time label that can be compared to the top of Figure 1 to show the point on the hydrograph when the sample was taken. Particle concentration is presented as number of particles per $\log_{10}$ of particle diameter per ml. The first two grab samples have the greatest concentration of particles (1.8×10^8/mL) and total particle concentration declined as the storm progressed (top of Figure 1). The 2 h grab sample has the greatest concentration (3.4×10^8/mL/μm) of the smallest particles (<0.8 μm in diameter) (bottom of Figure 1).

Table 1 Event summary

Event date	Monitoring site ID	Event rain (mm)	Cumulative precipitation* (mm)	Max intensity (mm/h)	Total flow volume (m^3)	Antecedent dry days (days)	Grab sample amount
10/16/2004	Site 1	11.9	19.6	24.4	20.0	215	8
	Site 2	21.8	29.5	30.5	254.3	215	12
10/26/2004	Site 1	61.5	171.7	36.6	704.2	6	12
	Site 2	48.3	168.4	27.4	711.0	6	12
	Site 3	45.2	161.4	30.5	131.7	6	14
12/5/2004	Site 1	14.2	191.8	12.2	53.2	8	12
	Site 2	17.0	168.9	12.2	250.6	8	12
	Site 3	14.7	182.0	12.2	24.9	8	12
1/7/2005	Site 1	156.0	603.9	33.5	1,847.6	2	12
	Site 2	287.0	197.2	33.5	4,392.6	2	12
	Site 3	202.2	640.3	51.8	758.1	2	12
2/10/2005	Site 1	68.8	683.9	27.4	449.6	8	12
	Site 2	78.2	839.3	27.4	1,011.2	8	12
	Site 3	52.1	703.6	24.4	176.8	8	12
3/18/2005	Site 2	5.1	1,106.6	3.0	51.6	5	12
	Site 3	2.8	968.2	3.0	4.1	5	5
4/28/2005	Site 2	32.8	1,200.4	27.4	522.0	31	10
	Site 3	29.7	1,053.0	42.7	104.9	31	7

*Refers to seasonal cumulative rainfall amount at a specific location. Usually the rainy season starts from late September in west Los Angeles area

Particle concentration does not monotonically decline during storms and we have generally observed two qualitative phenomena that impact the concentration. A short burst of new rainfall after the beginning of a storm can mobilize particles, particularly small particles, creating a surge in turbidity, TSS and particle concentration. When rainfall rapidly decreases, as in this case, less runoff is produced to dilute the particles, which allows their concentration to rise. After approximately 1 h the runoff decreases to nearly zero, which is responsible for the second peak particle concentration and turbidity at 2 h.

Particle number first flush (PNFF) ratio was developed to describe particle first flush (Li *et al.*, 2005). The PNFF ratio is defined as the normalized number of particles divided by normalized volume fraction at any point of the normalized runoff diagram. For example, $PNFF_{20} = 2.5$ 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. PNFF ratio was calculated for different particle sizes for all 18 storms. The average of $PNFF_{20}$ values for 18 storms for different particle size ranges is shown in Figure 2. Results presented in Figure 2 demonstrate that mean $PNFF_{20}$ values generally increase with increasing particle diameter. The smallest particles (0.5–2 μm) have the lowest $PNFF_{20}$ ratio of 1.4. Particles between 2 and 30 μm have $PNFF_{20}$ ratios between 1.5 and 2 and particles larger than 30 μm have $PNFF_{20}$ ratios larger than 2. On average, more than 40% of particles larger than 30 μm in diameter are contained in the first 20% of runoff volume. This shows that the early runoff has the greatest fraction of all particles, particularly larger particles, and suggests that treating the early portion of runoff will be more effective than treating a portion of the runoff throughout the storm.

Particle stability and natural coagulation of particles

Slow mixing experiments were conducted for seven rainfall events from October 26, 2004 to February 7, 2005. PSD was analyzed before starting each mixing experiment and the initial particle (0.5–400 μm) concentrations ranged from 1.76×10^7/ml to 5.64×10^7/ml with an average of 2.98×10^7/ml. PSD was measured periodically at different mixing times ranging from 0.5 h to 23 days for both mixed and unmixed samples.

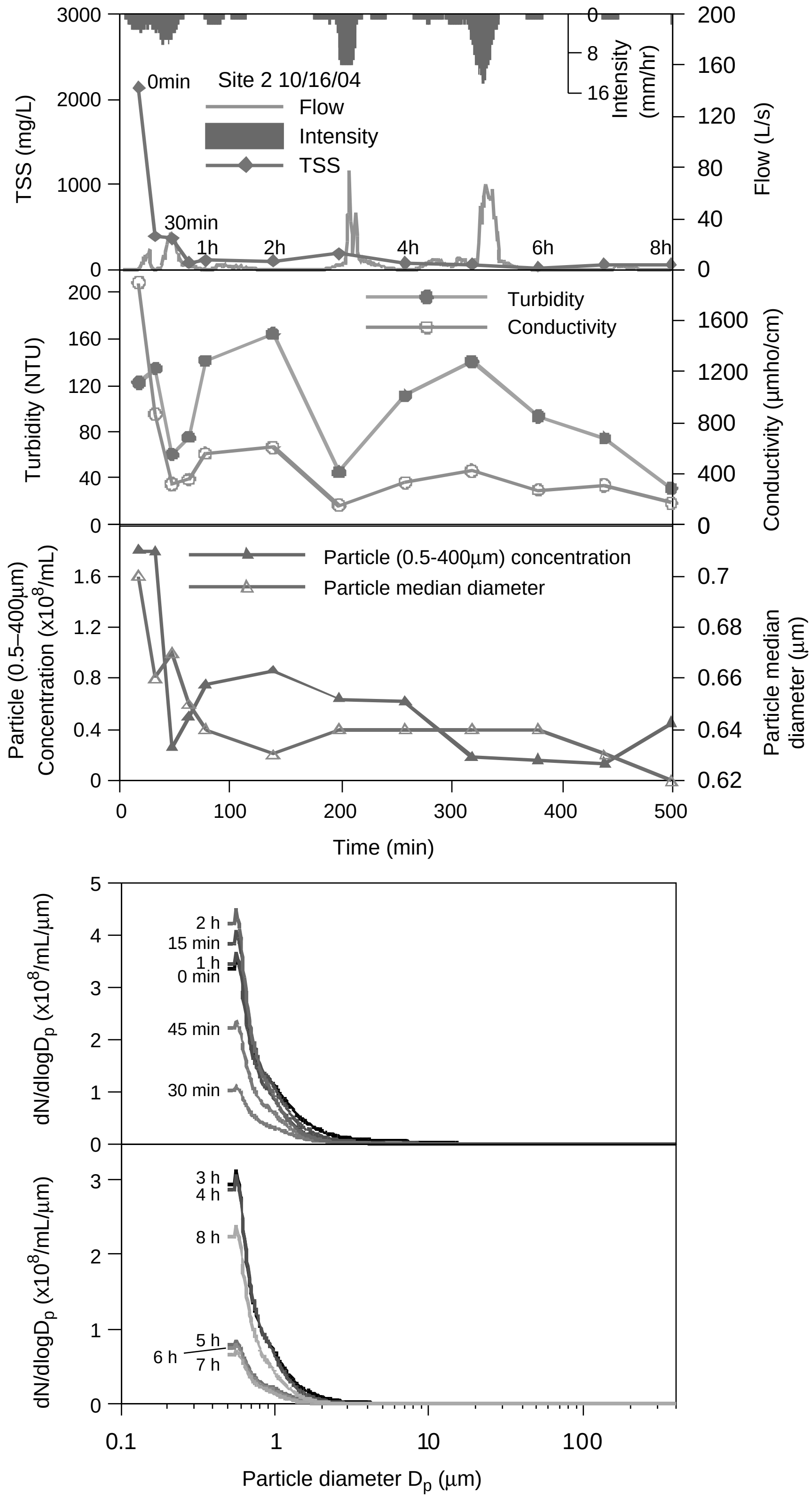

Figure 1 Hydrograph with TSS, turbidity, conductivity and PSD for site 2, event 10/16/04

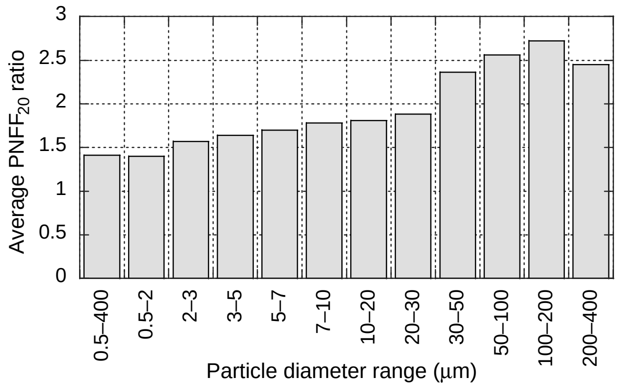

Figure 2 Average $PNFF_{20}$ ratio for different particle size ranges

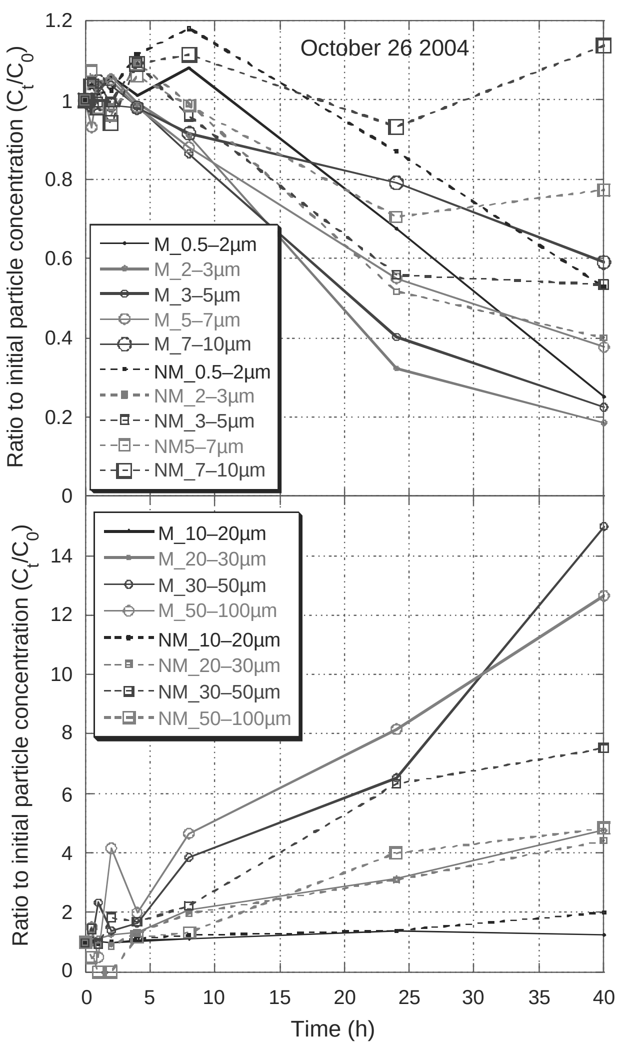

Figure 3 Particle size change with time for mixing and no-mixing experiments for first hour runoff sample of 10/26/04 from site 2

Particle concentration at different mixing times was normalized by dividing the particle concentration at specific times (e.g. 15 min, 1 h, 2 h, etc.) by the initial particle concentration. Figure 3 shows typical concentration ratio at different mixing times for both the mixed sample and the unmixed sample of October 26 2004. In the first 24 h, mixing greatly accelerated particle aggregation and decreased particle concentration by 32%, 68%, 60%, 45% of initial particle concentration for size ranges of 0.5–2 μm, 2–3 μm, 3–5 μm, and 5–7 μm, respectively. The corresponding unmixed sample particle concentrations decreased only 13%, 48%, 44% and 30% of initial concentration for the same particle size ranges. For particles in size ranges of 10–20 μm, 20–30 μm, 30–50 μm, and 50–100 μm, 24 h mixing increased particle concentration by 1.4 and 2.1, 5.5 and 7.2 times, respectively. After 24 h, the rate of particle aggregation of unmixed sample decreased, suggesting that an equilibrium PSD might exist.

Figure 4 is a box plot of the ratio of the 24-hour particle concentration to initial concentration for seven mixed and unmixed samples for different particle size ranges. On average, mixing decreased the concentration of particles less than 7 μm by 51%, while the concentrations of unmixed samples decreased by only 14%. For particles between 7 and 10 μm, mixing decreased particle concentration by 20% while the unmixed concentration increased by 120%. Particles number concentration decreased for particles smaller than 7 μm while particles number concentration for particles larger than 20 μm increased. In general, mixing greatly improved the particle aggregation compared with the same unmixed sample.

Slow mixing can greatly reduce small particle concentration without chemical addition, which might have implications in BMP designs. For example, a sedimentation tank can be designed to capture the first part of runoff volume until it fills to a maximum height of 3 m. This tank can gently be mixed for 24 h followed by 24 h sedimentation. Using the settling velocities of highway runoff samples measured by Li *et al.* (2006b), compared with unmixed sedimentation for a 24 h period, mixing can improve particle removal efficiency from 8% to 47%, from 32% to 81%, from 34% to 73%, from 42% to 81%, from 50% to 77%, from 65% to 68% for particles in size ranges of 0.5–2 μm,

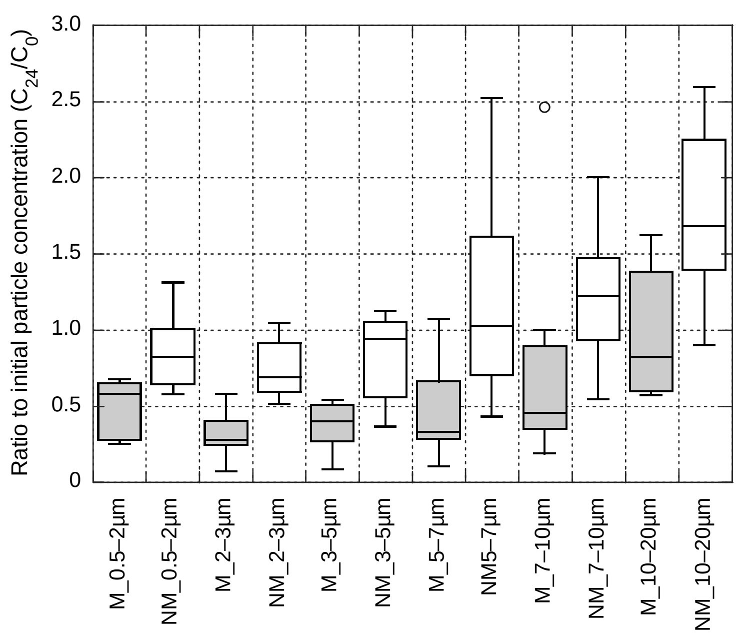

Figure 4 Particle concentration ratio to initial time for mixing and no-mixing samples at 24 h mixing time

2–3 μm, 3–5 μm, 5–7 μm, 7–10 μm, 10–20 μm, respectively. Under this proposed design, concentration of particles larger than 20 μm after 24 h mixing is much higher than unmixed condition. Therefore, it is expected to have more suspended particles larger than 20 μm in the sedimentation tank. If sand filtration is used after 24-hour mixing, overall particle removal efficiency will be greatly improved.

Conclusions

Particle size distribution (PSD) was measured for 200 grab samples from 18 storms during the 2004–2005 rainy season. Particles showed a strong first flush with larger particles having a greater first flush. On average, 28% of total number of particles between 0.5 and 2 μm, more than 30% of particles between 2 and 30 μm, and more than 40% of particles larger than 30 μm are carried in the first 20% of runoff volume. Mixing can effectively accelerate the particle aggregation process. After 24 h, mixing decreased particle concentration by 51% for particles less than 7 μm and the concentration of unmixed sample for the same size range decreased only 14%. For particles larger than 20 μm, mixing increased concentration by 6-fold while the unmixed concentration increased only 4.5 times. In general, mixing decreased smaller particle concentrations while increasing larger particle concentrations. Implications for settling were examined using specific settling tank design and a mixing regime. The results showed that slow mixing can improve sedimentation efficiency by more than 40% for particles less than 20 μm.

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