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# Characterization of suspended solids and particle-bound heavy metals in a first flush of highway runoff*

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**Abstract:** To investigate the dynamic characteristics of total suspended solids (TSS) and their particle-bound heavy metals in a first flush, the runoff sampling together with its flow rate measuring was conducted for three rainfall events at outfalls of highway in Shanghai from June to September 2007. Field samples were analyzed to determine the concentrations of TSS and particle-bound heavy metals, such as Zn, Pb, and Cu. Results show that the wash off behavior of TSS under varying runoff rate condition can be explained by different antecedent dry weather period (ADWP). Contribution of fine fraction (<45 μm) to TSS was generally higher than that of coarse fraction (>45 μm). When the runoff flow increased obviously, a significant contribution of the coarse fraction was observed for a certain rainfall events with long antecedent dry weather condition. The changes of total metals concentration and particle-bound metal concentrations were strongly dependent on the TSS variation. TSS was generally well correlated with most particulate-bound heavy metals. Of the heavy metals, the concentration of Zn was found considerably high and that of Pb was significantly low at North Zhongshan 2 Road, in Shanghai, China, but they are still within the range reported in the literature. Fluctuation of heavy metal contents in the coarse fraction during a first flush period was more significant compared with that in the fine fraction. The results will assist in the development of effective control strategies to minimize heavy metals and solids in highway runoff.

**Key words:** Heavy metals, Highway runoff, First flush, Suspended solids (SS)
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## INTRODUCTION

Non-point pollution has been identified as one of the leading sources of pollution in developed urban areas (US EPA, 1998; Drapper *et al*., 2000). Among many non-pollution sources, road/highway storm runoff is considered as an important source of micro-pollutants such as heavy metals (Barrett *et al*., 1998; Furumai *et al*., 2002). In China, 80% of the sediments in rivers and lakes are polluted by heavy metals (Zhou *et al*., 2004). Unlike organic contaminants, heavy metals do not degrade in the environment, and can exert both short- and long-term toxicity impacts by mass accumulation. During the wet weather period, the hazardous pollutants are washed off mainly at an early period (Smith *et al*., 2000). The initial runoff volume, where the pollutants concentration is substantially higher than that in the later period, is called "first flush".

For the last few decades, many researchers have focused on the issue of highway runoff pollution in developed countries (Hoffman *et al*., 1984; Krein and Schorer, 2000). In several field surveys, suspended solids (SS) in highway runoff have been found among the major pollutants since many micro-pollutants are attached to them (Sansalone and Buchberger, 1997a; Wu *et al*., 1998; Shinya *et al*., 2003). According to Furumai *et al*.(2002), high concentrations of heavy metals were observed at the early stage of runoff.

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Furthermore, Lau and Stenstrom (2005) reported that accumulating behavior of heavy metals was mostly dependent on particle sizes distribution. In fact, pollutant particle size distribution is crucial in stormwater treatment by some practices (e.g., wetlands, detention basins and retention pond). In Shanghai, the treatment technology for detention basins has already been used extensively for pollution control of Shanghai Suzhou Creek from initial stormwater runoff (Chen *et al*., 2004; Xu *et al*., 2005; Tan *et al*., 2006; Xu *et al*., 2006). The facilities are designed to improve runoff water quality and rely, to a large degree, upon pollution removal by settlement. Coarse particles will settle easily in detention basins, but fine particles may be discharged due to their slower settling velocities (Pitt *et al*., 1995). As a result, serious pollution still may be hardly avoided. Therefore, information on the flushing behavior of particles of different sizes and attached heavy metals in stormwater is very important to discuss possible control measures and develop potential treatment methods of runoff pollutants. However, there has been very limited field investigation so far about the dynamic behavior of SS and particle-bound heavy metals. Especially, little information is available for developing countries, including China.

The primary objective of this study is to investigate the characteristics of wash off behavior of TSS, fractional SS and their particle-associated heavy metals in a first flush of highway. The results will assist local governments to develop an effective control strategy of reducing stormwater micro-pollutants in the receiving waters when they were directly discharged.

## METHODS

### Study site

The study was conducted from June to September 2007 at North Zhongshan 2 Road in Shanghai, China. Runoff monitoring of nine rainfall events was carried out and three typical rainfall events among these were selected to analyze. Samples of rainfall events were collected at the outlet of drainage pipe. Service area of each drainpipe was about 400 $m^2$. In this highway, the average daily traffic volume was over 30000 vehicles per day.

### Sampling and analysis

Grab samples were taken manually and collected with a polypropylene bottle at the outlet of the drainage pipe. Sampling interval was 5 min in the first 30 min after the start of detectable runoff, and then increased to 15 min. The number of the samples collected depended on the rainfall intensity and duration. Time of sampling and runoff volume were recorded simultaneously. Flow rate was measured using a polypropylene bottle with 1 L or 4 L for smaller volumes and a polythene barrel with 10 L for higher volumes. Rainfall was recorded using an automatic rain gauge, which was positioned near the highway.

Water samples were taken to the laboratory after each event immediately. Samples were kept chilled in the lab and filtered within 24 h. TSS was analyzed according to APHA standard methods (APHA, AWWA, WEF, 1992). Each 50 ml dissolved fraction was immediately acidified with 2.5 ml of concentrated $HNO_3$. The solids retained on the filter were digested using a Questron microwave system to release particulate-bound metals into solution. 0.5 g of each particle fraction was weighed in the digestion vessel together with 10 ml concentrated $HNO_3$. Program microwave unit to heat samples to (160±4) °C in 10 min and then, at the second stage, to permit a slow rise to 165~170 °C in 10 min. After cooling, the digested samples were filtered, diluted to 50 ml and analyzed using Perkin-Elmer Optima 2100 Inductively Coupled Plasma Spectrometer (PerkinElmer Co., USA).

The runoff samples were sized into fine fraction of 0.45~45 μm and coarse fraction of 45~2200 μm using polyester sieve following the classification described previously (Furumai *et al*., 2001; Murakami *et al.*, 2004). The larger fractions were not found because they were better removed by existing street sweepers.

Laboratory quality control/quality assurance (QC/QA) procedures were followed, which were specified in Stormwater Monitoring Protocol Guidance Manual (Caltrans, 2000). Duplicate filtration and analysis were done for all samples per storm event sampled. Good precision of the whole procedure (less than 5% expressed as relative standard deviation) was achieved.

RESULTS AND DISCUSSION

**Temporal variation of TSS**

The characteristics of the monitored storm events are summarized in Table 1. These rainfall events represent different rainfall characteristics. In all the three stormwater, runoff samples were collected during the first flush period. Fig.1 shows the change of flow rate and TSS concentration along time during the storm events on June 23, July 20 and August 2, 2007, respectively. The high rainfall intensity was observed in the initial period of the events on June 23 and August 2, 2007. However, for the events on July 7, 2007, it was found in the following period.

As shown in Fig.1, the high degree variabilities of the flow rate and TSS were observed, particularly for the stormwater TSS, which indicated the variety of contaminant and the complex wash off dynamics of the micro-pollutions such as heavy metals (Han *et al*., 2006). In addition, the higher concentration of TSS at the early stage was found similar to that of the flow. Aryal *et al*.(2005) had similar observation during their research on SS in highway runoff. But occurrence time of the peak concentration for TSS was not entirely consistent with that of the peak runoff flow rate in all the three rainfall events. For the rainfall events on July 7, 2007, the concentration peak of TSS preceded the peak of runoff flow rate. Contrarily, for the other two rainfall events, the concentration peak of TSS dropped behind the peak of runoff flow rate or appeared almost at the same time with the peak of runoff flow rate. Generally, for all the storm events monitored, the highest TSS concentration always occurred within the first thirty minutes and declined as the storm progressed, no matter whether another flow peak existed or not. The increasing TSS concentration at the early period seemed reflecting the wash off of the deposited pollutants.

In this study, the rainfall characteristics and antecedent dry weather period (ADWP) could be responsible for this phenomena mentioned above. The magnitude of TSS concentration in the first flush was

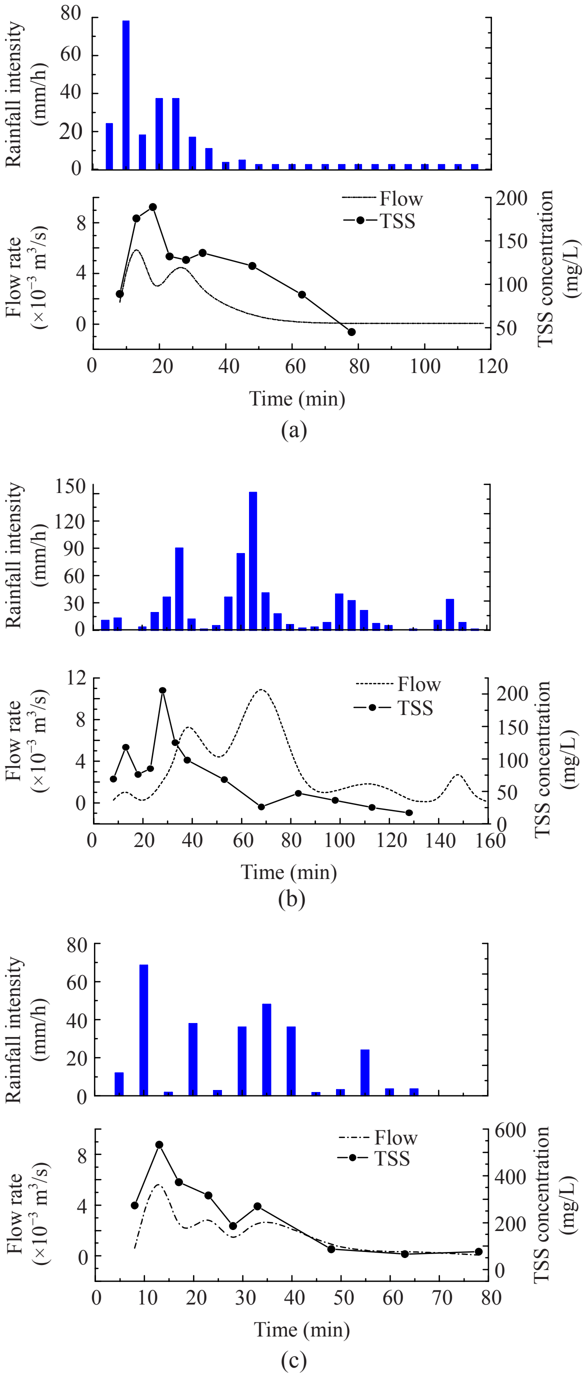


**Fig.1 Variation of TSS with flow rate along time during the storm events on (a) June 23; (b) July 7 and (c) August 2, 2007**

**Table 1 Characteristics of the observed rainfall events in 2007**

| Date of rain event | Rainfall depth (mm) | Rainfall duration (h) | Average rainfall intensity (mm/h) | Previous rainfall volume (mm) | Antecedent dry weather period (d) |
|---|---|---|---|---|---|
| June 23, 2007 | 19.9 | 1.2 | 16.6 | 35.9 | 1.28 |
| July 7, 2007 | 58.5 | 2.6 | 22.7 | 11.3 | 1.63 |
| Aug. 2, 2007 | 22.9 | 2.1 | 10.9 | 15.6 | 10.64 |

significantly correlated to the ADWP. The rainfall events with longer ADWP were more likely to result in higher TSS concentration. The rainfall event on August 2, 2007 was preceded by the longest dry weather period compared with other rainfall events in this study as shown in Table 1. Moreover, its rainfall intensity could be sufficient to flush out the deposited mass on the surface quickly. Therefore, it produced the highest TSS concentration in the first flush among the three rainfall events. When rainfall rapidly decreased, less runoff was produced to dilute the residual particles. However, high TSS concentration still could be measured. Comparatively, the rainfall events on June 23 and July 7, 2007 had relatively short ADWP, which provided a less buildup mass in the same street-sweeping conditions. Consequently, shorter ADWP caused lower peak concentration of TSS. After the peak, the TSS concentration rapidly declined to a low concentration even at a high peak flow since there were no enough particles available for wash off to continue. Other investigators also observed that the ADWP and rainfall intensity significantly affected the TSS concentration of runoff from highway (Lee *et al*., 2002; Li *et al*., 2007).

**Variation of fractional SS in runoff**

The Federal Highway Administration of USA reported that 70% of the particles from highway runoff were smaller than 45 μm (Kobriger, 1984). Similarly, Furumai *et al*.(2001) reported that the fine particles smaller than 45 μm accounted for almost half in mass of the TSS in runoff. Hence, in this study, the particle size was fractionated considering 45 μm as a borderline following the previous studies. Fig.2 shows the size distribution of fine (<45 μm) and coarse particles (>45 μm) in runoff samples during the first 30 min after the runoff began on June 23, July 7 and August 2, 2007. The dotted line denoted the flow rate during the sampling period. From Fig.2, it showed that wash off behaviors of fine and coarse fractions were various even for the same rainfall events. In the three rainfall events, both fine fraction and coarse fraction fluctuated significantly with time, and the coarse fraction had a greater fluctuation. Contribution of fine fraction to TSS was generally greater than that of coarse fraction. Especially for the rainfall events on June 23 and July 7, 2007, over 70% of the fine fraction was contained in TSS. On the contrary, in the rainfall event on August 2, 2007 a significant contribution of the coarse fraction was observed when the flow increased obviously. However, a similar flow increase on June 23, 2007 did not cause a significant rise of coarse fraction contribution.

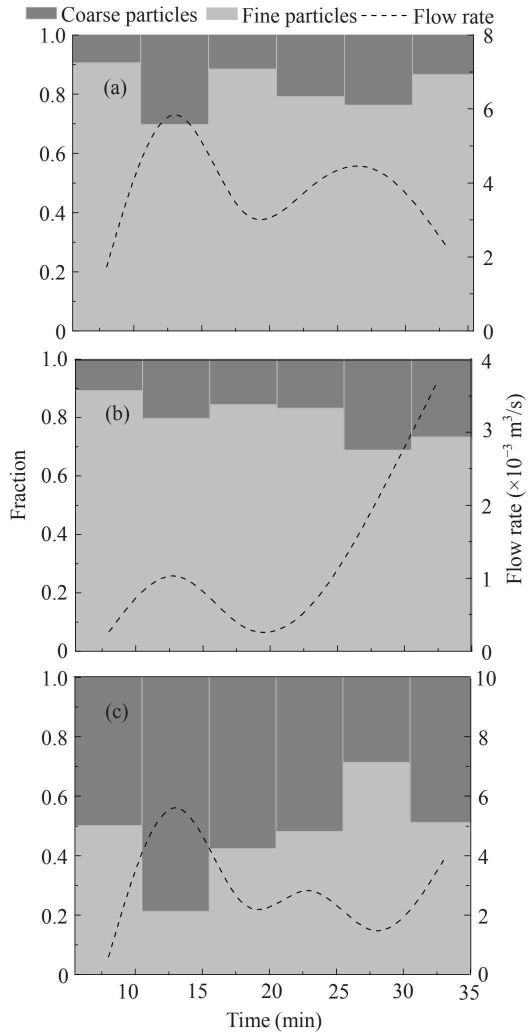


**Fig.2 Fractional contribution by weight of fine and coarse particle along time during the storm events on (a) June 23; (b) July 7 and (c) August 2, 2007**

These results may be largely attributed to the difference of the fine/coarse ratio in particulate matters. It had been demonstrated in previous researches that the fine fraction in TSS accounted for major portion within the low range TSS. However, things went opposite direction when the TSS concentration was high, i.e., coarse fraction showed power growth relation with respect to the TSS concentration (Aryal *et al*., 2005). That is to say, the coarse fraction increased with the growth of TSS. Furthermore, TSS loads were largely influenced by ADWP. Hence, the antecedent condition may result in a variation in the fine/coarse ratio despite a similar flow rate. As for the August 2 event, the longer ADWP resulted in a more buildup mass, which in turn rendered a higher TSS load. Accordingly, coarse particulate could account for a significant portion of total particulate matter load. When rainfall increased rapidly, a short burst of rainfall could form sufficient runoff to mobile particulate matters, creating a surge in TSS, particularly for coarse particle. So a significant contribution of the coarse fraction was observed in the rainfall event on August 2, 2007. In contrast, fine particulate in TSS accounted for larger portion due to the relatively short ADWP for the rainfall event on July 7 and June 23, 2007. At the beginning of rainfall event, TSS built up on this surface were easily washed off by runoff since flushing of roadway surfaces by runoff was more effective in mobilizing the very fine fraction. In general, the difference of the fine/coarse particle mass ratio among the three typical rainfall events is mainly dependent on the antecedent conditions (antecedent rainfall, ADWP, etc.).

## Concentration profile in total and particle-bound heavy metals

There is a general belief that many stormwater pollutants are adsorbed on the surface of SS, and previous researchers have correlated heavy metals to TSS in highway stormwater runoff (Sansalone and Buchberger, 1997a; Shinya *et al*., 2000).

Zn, Cu, Cr, Cd, and Pb were chosen for this study because they are of particular concern due to their prevalence, toxicity, and persistence in the environment. However, of the heavy metals, Cu and Zn were routinely detected, Pb was usually detected, Cr was sporadically detected, and Cd was never detected in all the events. So this study focused on Zn, Cu and Pb.

Fig.3 shows the dynamic behavior of the total (T-Zn, T-Cu, T-Pb) and particle-bound (P-Zn, P-Cu, P-Pb) heavy metal concentrations as a function of time. As shown, the peak concentration of heavy metals appeared in the third, fifth and second sample for the three rainfall events, respectively. Especially for the rainfall event on August 2, 2007 the highest concentration appeared in the second sample and then gradually decreased, showing strong first flushing effects. Rain pattern may be an important reason for the significant first flush. The total and particle-bound concentrations of metals (e.g., Zn, Cu) were highly variable from one to another rainfall event; in contrast, Pb concentrations were less fluctuating. The dynamic behavior of total and particle-bound heavy metals concentrations can be well understood from Fig.3 that shows their relationship with TSS. The changes of the total concentration and particle-bound concentration were strongly dependent on the TSS variation. Consequently, the concentration profiles of heavy metals in total and particle showed a similar trend to

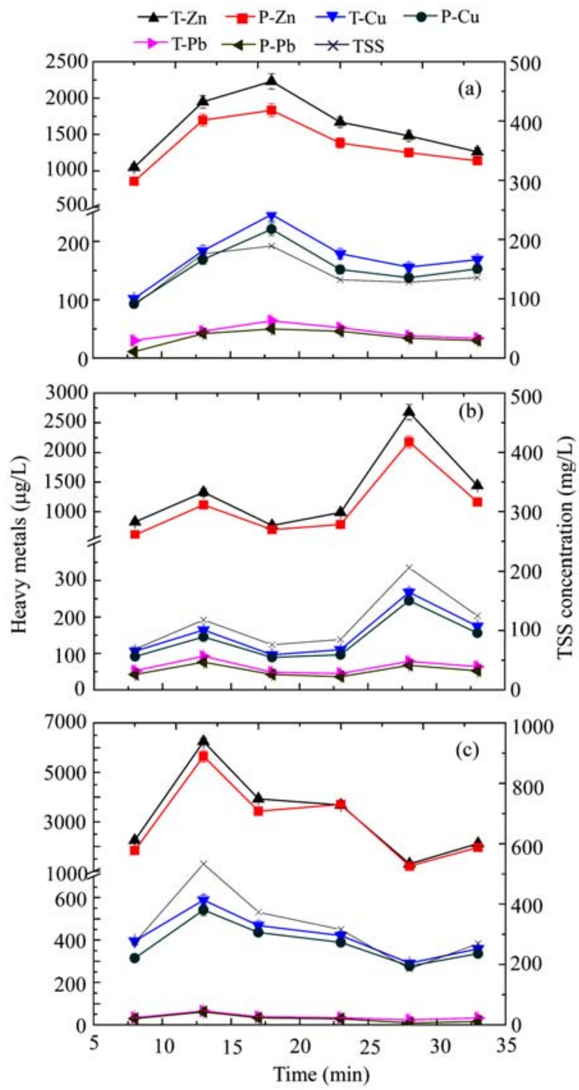


**Fig.3 Dynamic behavior of total and particle-bound heavy metals concentration during the first flush in the storm events on (a) June 23; (b) July 7 and (c) August 2, 2007**

corresponding TSS profiles. For the rainfall events on June 23 and on August 2, the concentration profiles of heavy metals were similar to each other but some difference was still observed in the event on August 2, which had a long ADWP as already discussed. As expected, the highest copper and zinc concentrations were found in the rainfall event on August 2. Moreover, the concentrations of copper and zinc were substantially higher during the early portion of the runoff. Comparatively, the rainfall event on June 23 was a smaller event (19.9 mm) with a relatively short ADWP, which limited maximum pollutants concentration.

Table 2 gives the maximum and minimum heavy metals concentration values from this study and other research. Generally the concentration levels follow the order of Zn>Cu>Pb. The zinc concentration was always greater than others, and was usually significantly greater. Although the Pb concentrations were in the range as shown in Table 2, they were significantly lower than the values reported by Sansalone and Buchberger (1997b) and Drapper *et al*.(2000). The Pb levels were much closer to the recent data reported by Tuccillo (2006). It was visible that the time data obtained was different. The Pb concentrations were much closer to the recent data. The low concentration of Pb in highway runoff of Shanghai might be due to the introduction of lead-free gasoline in China since 2000. However, Pb, which is still generated by traffic activities despite the phase out of leaded gasoline, is considered of the greatest concern. The Zn concentrations were considerably high at North Zhongshan 2 Road, Shanghai, but they were still within the range reported in certain literature (Table 2). Previous studies indicated that tire wear, diesel vehicle exhaust and the corrosion of galvanized safety barriers are the major sources for Zn in highway runoff (Barrett *et al*., 1998; Davis *et al*., 2001; Brown and Peake, 2006). Apparently, for China, the high zinc concentration primarily results from the use of Zn additives in formulating the oil to provide wear protection for tires, which is released by tire wear and subsequent water contact (Gan *et al*., 2008). Another important factor that should not be ignored is the heavy lorries traffic. Most of the heavy lorries are equipped with diesel motor. More pollutants such as particulate-bound Zn can be discharged with their exhaust gas for the quality of diesel oil under the current technical conditions in China. Besides, perhaps the gaps on road management and maintenance between China and those developed countries also made the great difference. Generally, Cu concentrations were similar to what has been reported by other researchers but only a few samples were available (Berbee *et al*., 1999; Furumai *et al*., 2004). Brake wear and fluid leakage are major source of copper. Brake pad material can contain copper, contributing the metal to environment during brake wear. An estimate of copper releasing from automobile brake abrasion has been given as 1.5 mg/(km·vehicle) (Davis *et al*., 2001). Additionally, it should be pointed out that atmosphere deposition has been reconsidered to be another very important source of copper (Sabin *et al*., 2006).

Furthermore, based on the obtained data, the results presented in Fig.3 indicated that the concentrations of Pb and Zn mostly or frequently exceeded the maximum values permitted by the Discharge Standard of Pollutants For Municipal Wastewater Treatment Plant (GB18918-2002, 2002). Moreover, judged by US EPA water standard for surface water discharge (US EPA, 1994), the concentrations of Cu and Zn in the runoff were found to be far exceeding surface water discharge concentrations limits for the adjacent receiving waters except for Pb. The results showed that highway runoff was badly polluted, especially for Zn. Therefore, Zn and Cu should be the primary targets of reduction by control strategies.

**Table 2 Comparison of heavy metals concentrations in highway runoff from various studies**

| Sources | Site | Cu (μg/L) | Zn (μg/L) | Pb (μg/L) | ADT* | Pavement materials |
|---|---|---|---|---|---|---|
| Sansalone and Buchberger (1997a; 1997b) | Highway | 43~325 | 459~15244 | 31~97 | 150000 | Asphalt |
| Drapper *et al.*(2000) | Highway | 30~90 | 175~440 | 50~250 | 6000~50000 | Asphalt |
| Tuccillo (2006) | Highway | 3~65 | 7~210 | 5~38 | — | — |
| This study | Highway | 96~588 | 770~6250 | 13~66 | 30000 | Asphalt concrete |

*ADT: average daily traffic

### Change of particle-bound heavy metals content in fractional SS

Particle-bound heavy metals content in the fine and coarse fractions for the three rainfall events are shown in Fig.4. From the figure, it was noticed that the fluctuation of the particle-bound heavy metals content in the coarse fraction was significant in the first flush period, while the particle-bound heavy metals content in the fine fraction was less fluctuating. This indicated that the runoff behavior of particle-bound heavy metals could be considered depending on particle size distribution. What is more, the smaller particle fraction had higher solid-phase heavy metals content in most cases. According to the result of Sansalone and Buchberger (1997a), the highest concentrations of Zn, Pb, and Cu in the size range of smaller than 45 μm were mostly found. The result was consistent with that in this study. Together, these confirmed that small silt-sized particles could be a primary vehicle for heavy metals transport in highway runoff.

Because particulate-bound heavy metals especially those bound by fine fraction dominate the heavy metals content in highway runoff, it is suggested that runoff treatment practices in Shanghai should be designed to improve retention of particulates removal. Best management practices (e.g., infiltration basin, grass swale, rain garden) with good performances on particulate removal may be favorable choices in highway runoff.

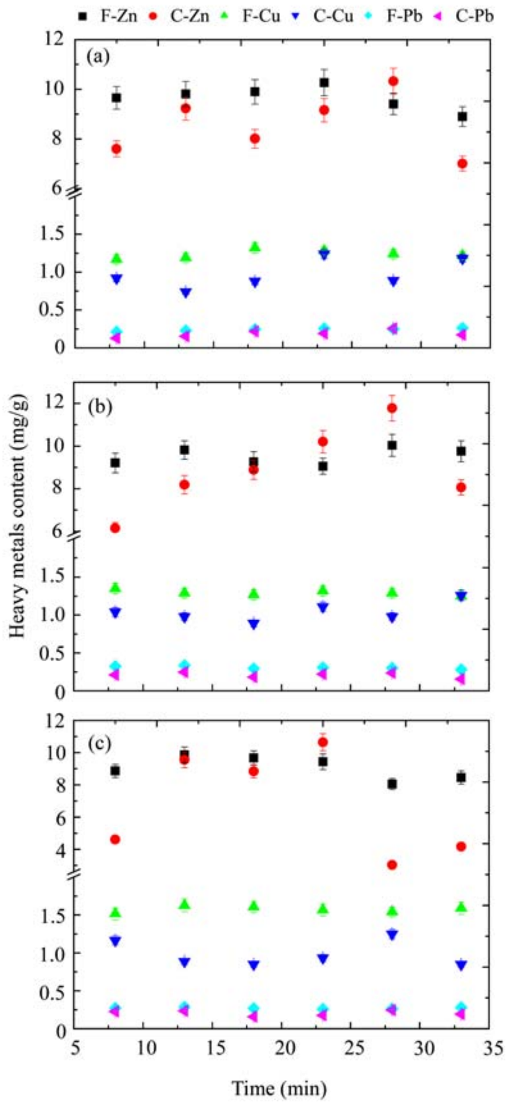


**Fig.4 Variation of particle-bound heavy metals content in fine and coarse fractions during the first flush in the storm events on (a) June 23; (b) July 7 and (c) August 2, 2007**

## CONCLUSION

The higher concentration of TSS at the early stage was found to be accompanied by a larger flow. The occurrence time of the TSS concentration peak and interval between the TSS concentration peak and the flow peak were found to be dependent on the rainfall characteristics and ADWP.

Contribution of fine fraction to TSS was generally higher than that of coarse fraction, except in the obviously increased runoff flow for certain rainfall events with long antecedent dry weather condition, a significant contribution of the coarse fraction was observed. This might be explained by the different wash off behavior of fine and coarse particles.

Heavy metals concentration profiles in total and fractional SS showed similar tendency to the corresponding TSS concentration profiles. As a whole, heavy metals content in fine fraction was higher compared with that of coarse fraction. Heavy metals content enrichment in fine fraction indicated the importance of fine particles runoff behavior.

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