Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2014.942703

56 (2015) 655–664
October

Taylor & Francis
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Particle size distributions in combined sewer overflows in a high-intensity urban catchment in Shanghai, China

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Received 26 March 2014; Accepted 24 June 2014

ABSTRACT

To determine the dissolved-phase chemicals and solid-phase particulate matter (PM) in a combined sewer overflow (CSO), the event mean concentrations of typical particulate-band and dissolved-band contaminants were analyzed. Particulate-band chemical oxygen demand and total phosphorous formed the majority of particles, while only around 50% of the total nitrogen was found in the particulate-band. Particle size distributions (PSDs) for PM in 49 CSO samples collected during seven rainfall events, and 18 run-off samples collected during two rainfall events, were analyzed. Variation in PSDs in both CSO and dry weather flow samples occurred with sample storage time. Mean values of particle size in CSO samples were not significantly different from median values for particles within the size range from 0.41 to 3,080 μm, at a 95% confidence level. Values for D50 and D90 for PSDs of PM in both CSO and run-off samples were higher than for previous studies. It was caused by high flow rates during the operation of storm water lifting pumps. The first flush of particles in different size ranges was quantitatively evaluated using PSDs and total suspended solids in each sample. Given the prevalence of CSOs in China and other countries, knowledge of the PSD, and its variation with time, facilitates both the design of CSO treatment procedures and optimization of wastewater treatment plants; it also provides important input for modeling water treatment operations.

Keywords: Combined sewer overflow; Particulate matter-first-flush; Particle size distribution; Pump drainage system; Run-off; Sample storage time

1. Introduction

Combined sewer overflows (CSOs) have significant impacts on the water quality of most old town centers in cities around the world. A complex mixture of dissolved-phase chemicals and solid-phase particulate matter (PM) occurs in CSOs. Event mean concentration (EMC), first flush, and settling processes for typical contaminants are well characterized [1–3]. Hence, in recent years, priority pollutants and micropollutants have been the focus of research [4,5].

PM is a main pollutant in CSOs; it is also a key index used in urban drainage maintenance. Since PM provides a reactive surface for adsorption of aqueous constituents, most of the typical contaminants, metals, priority pollutants, and micropollutants occur in the particulate-band in CSOs [4,5]. Thus, PM transports most of the contaminants in CSOs. Knowledge of its physicochemical characteristics will enhance our

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understanding of contaminant transport in combined sewer systems.

Particle size distributions (PSDs) for PM reflect both PM transport and fate. They provide more precise size information than PM classes or PM aggregate indices. PSDs for run-off and CSOs from urban areas are reported in many studies [6–8]. In addition, knowledge of PSDs is useful for designing water treatment facilities, especially for PM separation using a vortex separator.

First flush is an important phenomenon in CSO pollution; it affects the characteristics of PM transport. Several different methods for first flush analysis are used [1,9,10]. Although new methods were recently proposed [11,12], usually traditional methods are used for first flush analysis [13–15]. The Mass First Flush Ratio (MFFR), which divides mass by the cumulative run-off volume at a defined point, is most commonly used for a first flush detection [16–18]. To evaluate the MFFR, the concentration strengths of the first flush for different sized particles were quantitatively evaluated using PSDs and total suspended solids (TSS). These data are the characteristics of PM transportation for different particle size ranges.

In this study, the dissolved-band and particulate-band of contaminants in a CSO from a sewer system in urban Shanghai were investigated. Moreover, variation in PSD over time was analyzed, and its relationship to sample storage time was evaluated. First flush of different particle sizes also was determined. Understanding the characteristics of particles in the CSO is relevant to research on transportation of typical containments and other pollutants in wastewater, and facilitates design and optimization of water treatment plants.

2. Materials and methods

2.1. Description of the pump-lift drainage system

The Anshan experimental site is located in Shanghai, a metropolitan city in southern China (Fig. 1). Statistical analysis of its 30-year precipitation record (1971–2000) indicates that the average annual precipitation is 1,184.4 mm/year; while the average annual number of rainy days is 122.2.

The Anshan drainage system is a combined sewer system with a high intensity urban catchment, comprising mixed residential and commercial land use zones (Fig. 1). Its total area is 130 ha, with a residential population of 48,100. The pump station has four stormwater lifting pumps and four interception pumps. The nameplate flow rates for two of the

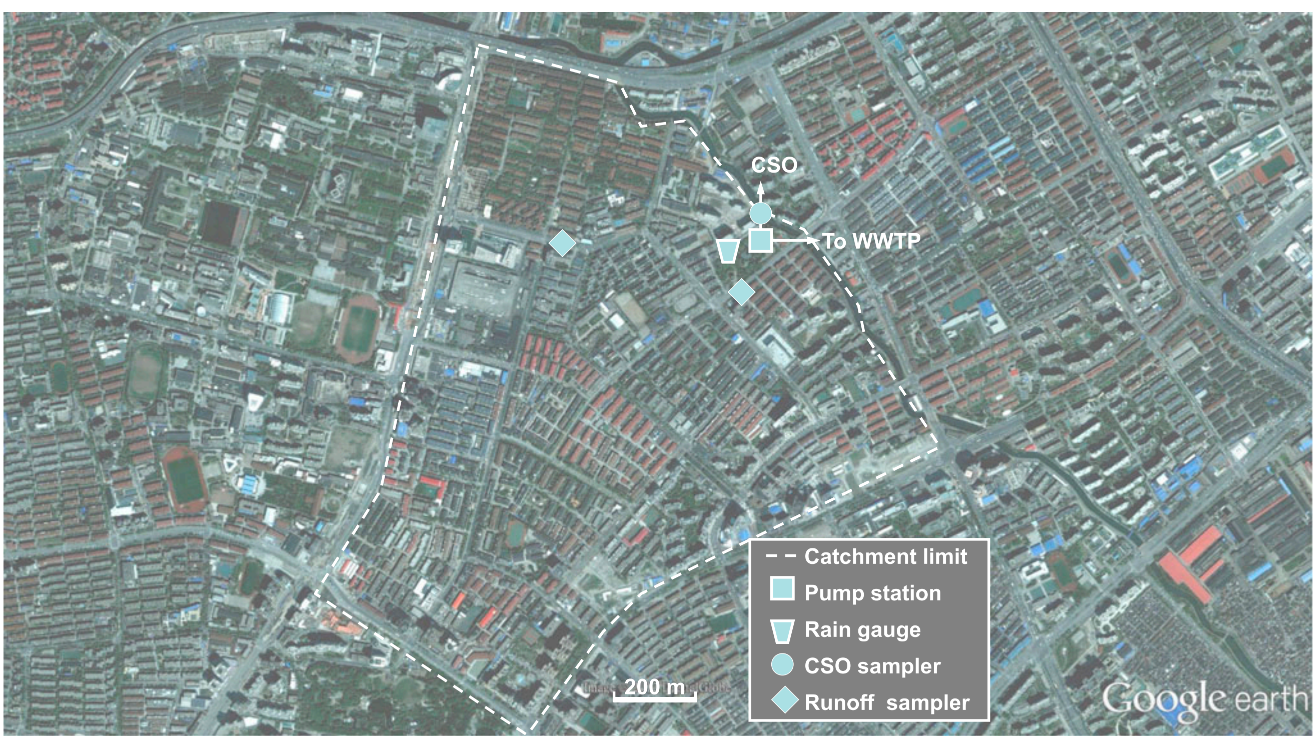

Fig. 1. The Anshan experimental site (Shanghai, China), showing catchment area and location of pump station and samplers.

Table 1
Characteristics of seven rainfall events between June and September 2013

Event date	Antecedent dry day (d)	Event rainfall (mm)	Run-off volume (m^3)	Overflow volume (m^3)	Max intensity (mm/ 5 min)	Rainfall duration (min)	CSO duration (min)	Max overflow flow rate (m^3/s)	Average overflow flow rate (m^3/s)
25 June 2013	35	28.4	25,800	23,800	8.7	240	80	5.1	4.6
05 July 2013	9	29.3	26,700	15,500	5.5	590	75	2.8	2.8
01 August 2013	3**	32.7	29,800	20,000	5.1	255	75	5.1	4.2
04 August 2013	3	44.9	40,900	28,700	8.4	55	80	7.9	5.6
25 August 2013*	21	41.0	37,300	21,600	10.8	80	90	5.6	4.2
13 September 2013*	18	86.9	79,100	60,700	10.2	585	170	10.2	6.0
21 September 2013	8	15.6	14,200	10,700	2.4	255	55	2.8	2.5

*Run-off was sampled simultaneously.
**Because of equipment failure, the CSO for the rainfall event on 29 July 2013 was not successfully sampled.

stormwater lifting pumps are 2.8 m^3/s, while the other two are 2.3 m^3/s. The nameplate flow rates for two of the interception pumps are 0.40 m^3/s, while the other two are 0.28 m^3/s.

2.2. Field sampling

An automatic water sampler (ISCO 6712, ISCO Inc., NE, USA) was installed at the pump station at the Anshan experimental site for dry and wet weather periods. Wet weather flow data were obtained from the SCADA system for the pump station. A rain gage at the pump station provided rainfall data.

Sampling was carried out between June and September 2013. CSO samples were collected automatically at the pump station, while run-off was sampled manually at two sites (Fig. 1). CSO and run-off samples were taken at 15 min intervals during rainfall events. Dry weather flow (DWF) samples were taken at 120 min intervals. Characteristics of seven rainfall events at the Anshan experimental site were recorded during the study period (Table 1).

2.3. Physical and chemical analyses

The automatic sampler was set to collect 1,000 ml of water in a polyethylene bottle per sample; samples were recovered immediately following a rain event or very early the next morning (when rain occurred at night) and transported to the laboratory for analysis. Samples were tested for TSS, chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) using standard methods [19]. Samples were filtered through a glass fiber filter (0.45 μm) and these filtrates were tested for dissolved chemical oxygen demand (DCOD), total dissolved nitrogen (TDN), and total dissolved phosphorus (TDP). Particulate chemical oxygen demand (PCOD), total particulate nitrogen (TPN), and total particulate phosphorus (TPP) also were calculated using the difference between total pollutant and dissolved-band pollutant concentrations. When not immediately analyzed, samples were kept refrigerated, and processed within 24 h.

A Malvern Mastersizer 3000 (Malvern Instruments Ltd, UK) with a particle size resolution from 0.01 to 3 500 μm was used in this study to determine PSD within 6 h of collection. Replicate subsamples were analyzed. PSD output was converted to gravimetric concentration using the values for TSS and particle density.

3. Results and discussion

3.1. Particulate and dissolved contaminants

Listed below are the EMCs for typical contaminates in CSO for each rainfall event studied (Table 2).

In CSO samples, the particulate band comprised more than 81% of the COD, as well as more than 75% of the TP. However, only 39–62% of the TN occurred in the particulate-band.

To investigate variation of the particulate and dissolved proportion of contaminants in discharge processes in the CSO, the COD, TN, and TP in each fraction were plotted against time (Fig. 2).

Clearly, concentrations of the particulate-band COD and TP in samples taken within the initial 10 min of the CSO were significantly higher than in subsequent samples (Fig. 2). In all samples, the particulate-band COD accounted for around 82–93% of the total COD; while the particulate-band TP accounted for 62–83% of the total TP. The particulate-band TN accounted for 52 ± 13% of the total TN in the first sample, dropping to 45 ± 13% in subsequent samples.

Table 2
Event mean concentrations (mg/L) of particulate and dissolved contaminants in the combined sewer overflow

Event	TSS	COD	DCOD	PCOD	TN	TDN	TPN	TP	TDP	TPP
25 June 2013	1,184	1,078	–	–	39.8	–	–	6.88	1.15	5.73
05 July 2013	715	913	–	–	29.4	–	–	4.38	0.7	3.68
01 August 2013	522	454	87	367	21.1	12.5	8.6	3.74	0.93	2.81
04 August 2013	461	452	39	413	21.4	8.0	13.4	2.86	0.45	2.41
25 August 2013	618	721	114	607	33.9	20.7	13.2	4.68	1.47	3.21
13 September 2013	581	457	35	422	19.3	10.1	9.2	2.76	0.59	2.17
21 September 2013	670	965	86	879	46	21.8	24.2	5.31	1.62	3.69

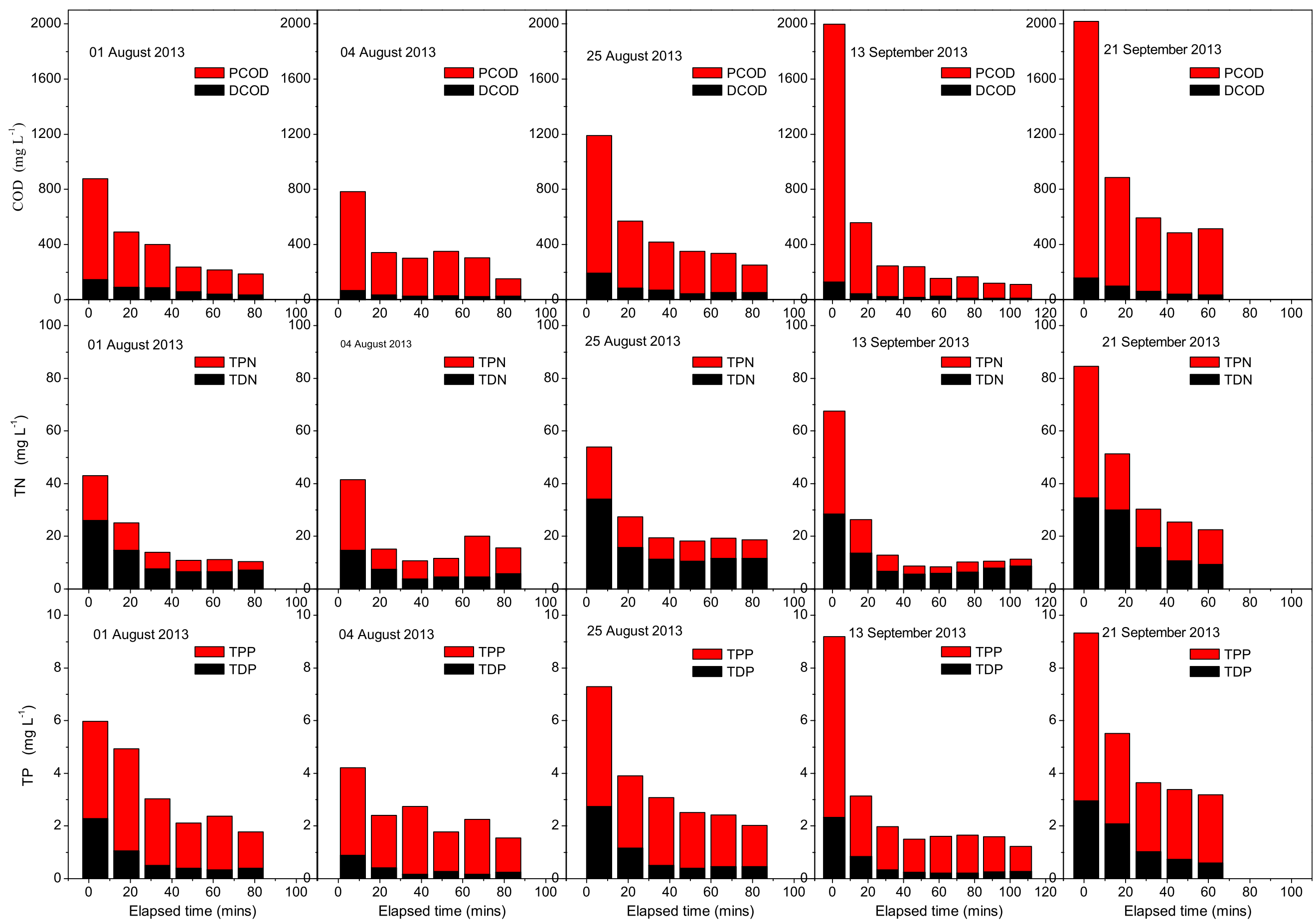

Fig. 2. Particulate-band and dissolved-band COD, TN, and TP in CSO samples.

Source apportionment for TN in CSO samples at this experimental site indicated that dissolved ammonium as nitrogen (NH_4^+–N) accounted for a relatively large proportion of the TN in the CSO samples, mainly occurring in the dissolved-band [20].

3.2. Variation in PSD with sample storage time

Particle aggregation or dissolution may affect PSD with increasing sample storage time. To quantify changes in PSD with sample storage time, five grab samples of CSO taken on 21 September 2013 and one sample of DWF taken on 19 September 2013 were analyzed at different time points, using the Malvern Mastersizer 3000 after gentle inversion (Fig. 3). Samples were kept at 4℃ in the refrigerator between analyses. Variation in PSD with time for these six samples is shown in Fig. 3.

In general, particle size increased with increasing storage time for all samples. A marked change

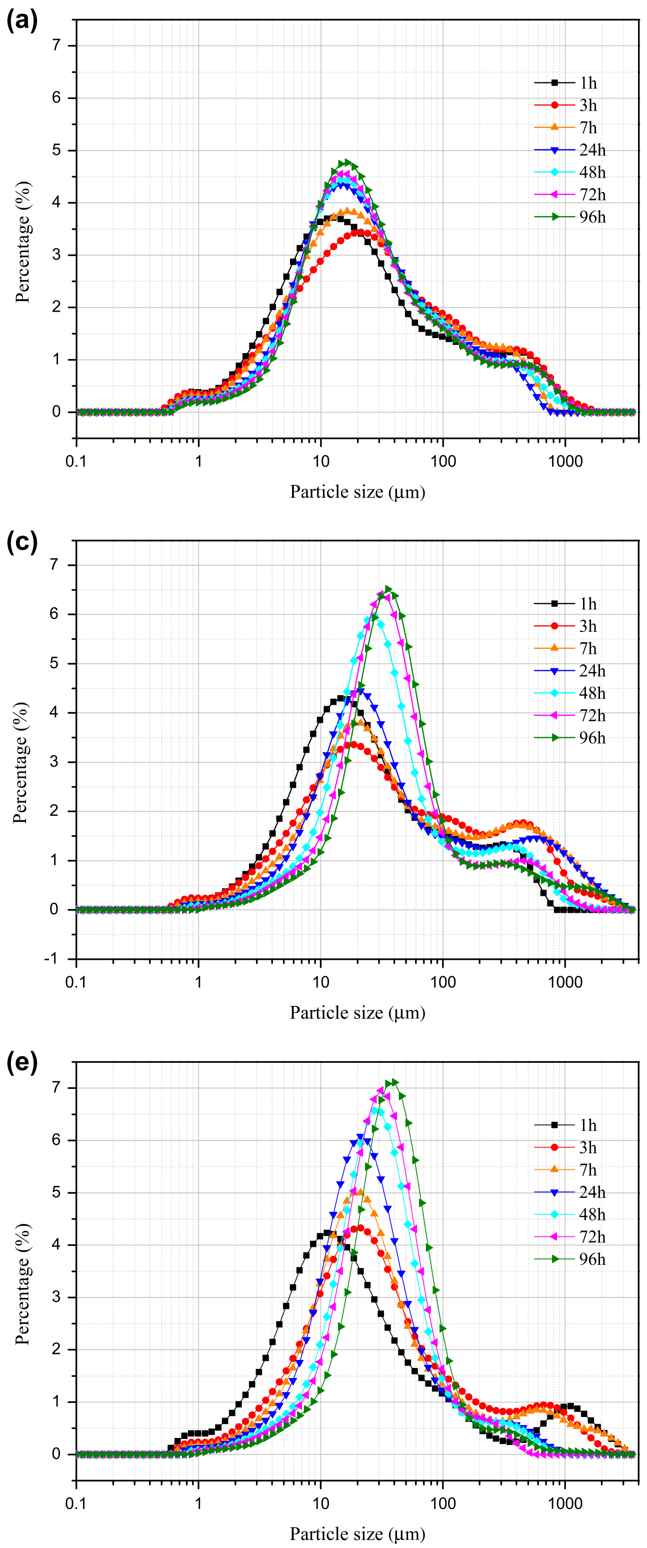

Fig. 3. PSDs for CSO samples and one DWF sample with increasing storage time of samples. CSO samples shown here were collected at various times following the start of the CSO discharge (1, 16, 31, 46, and 61 min).

occurred at around 24 h, with a change in peak particle size and shape of the distribution curve, suggesting that aggregation occurred mainly before this time point. A *t*-test was used to compare the PSD before 24 h and the PSD post 24 h. The *t*-test indicated that the PSD before 24 h was significantly different from the PSD post 24 h at a 95% confidence level. Post 24 h, the DWF sample appeared to change least, suggesting there was a balance between aggregation and dissolution processes at this point.

In all CSO samples, the proportion of smaller particles (<10 μm) decreased significantly over time. The proportion of small particles (<10 μm) in the DWF sample showed a similar trend, changing from a bimodal to unimodal distribution with larger particle size with time. Particles with a size range from 10 to 100 μm, in both CSO and DWF samples, increased in size with increasing storage time. Less change appeared to occur for these larger particles in CSO samples for storage times greater than 72 h.

Particles larger than 200 μm in both CSO and DWF samples do not show a clear trend. This may reflect different initial distributions of particles in samples. The rapid increase in particles between 10 and 100 μm in size, and the decrease in smaller particles (<10 μm) likely reflects naturally occurring coagulation and flocculation processes.

The influence of sample storage time on PSDs of PM is supported by these data. Hence, PSDs of PM in samples must be analyzed as soon as possible. However, a certain time is needed for both sampling on site and transporting the samples to the laboratory. For all the samples, it is difficult to do PSD analysis immediately after collection. Thus, considering the difficulties in practical application and experiment, the PSDs of PM should be analyzed preferably within 6 h of collection at least.

3.3. *PSDs in combined sewer overflow samples for different rainfall events*

Seven overflow events related to rainfall were monitored between June and September 2013. Between 7 and 14 samples were taken during each event. Thus, 49 samples from six of these events were analyzed to determine their PSD (not including 1 August 2013 samples). The mean and median PSDs for all samples are plotted against data for all samples (Fig. 4).

The minimum particle size in CSO samples was 0.41 μm, while the maximum particle size was 3,080 μm. A t-test was used to compare the mean proportion and the median proportion of particles with sizes in the range from 0.41 to 3,080 μm. The t-test indicated that the mean particle size was not significantly different from the median value at a 95% confidence level.

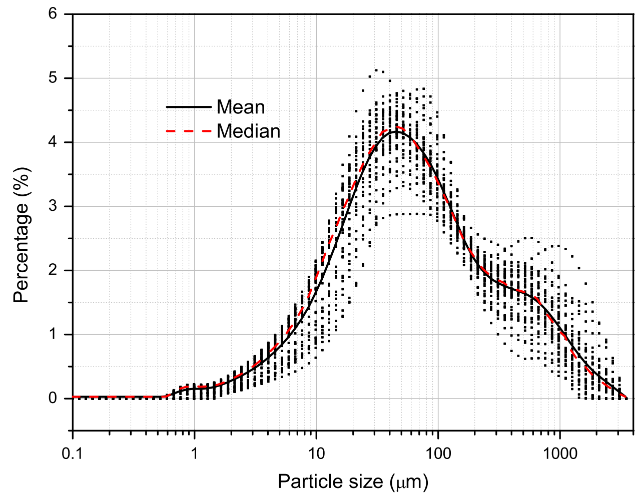

Fig. 4. PSDs of PM in combined sewer overflow samples collected between June and September 2013.

Distributions for particles in the size range from 0.41 to 1 μm were consistent for all samples. Distributions for particles between 1 and 3,080 μm in size, showed large variations among samples. In particular, for particles with size ranges between 20–60 μm and 200–3,080 μm, there were significant differences between rainfall events ($p < 0.05$). Table 3 shows values for D10, D50, and D90 of PSD for PM in CSO and run-off grab samples for each rainfall event.

The mean D50 for PSDs of PM in CSO grab samples from different rainfall events ranged from 52.1 to 83.2 μm (Table 3). This range was higher than values reported in a combined sewer system in the Liguori catchment, Cosenza, Italy [8].

For rainfall events on 25 August 2013 and 13 September 2013, run-off from two sites in the Anshan catchment area was sampled, yielding 18 run-off samples. The D50 of PSD for PM in run-off samples ranged from 25.2 to 45.8 μm, while the D90 ranged from 256 to 1,060 μm (Table 3). These ranges are also larger than for PSDs reported for in run-off samples in previous studies [6,7].

It should be noted that the mean D10, D50, and D90 of PSD for PM in run-off samples were lower than for those for CSO samples. The different character of the PSDs suggests that the large particles in run-off samples may not be the main contributor to the large particles in CSO samples. Eroded sediments and aggregated particles in the sewer system may be a major component of the large particles in CSO samples. In a pump-lift combined sewer system, the high flow rate caused by stormwater lifting stations could lead to serious sediment erosion.

Table 3
Values for D10, D50, and D90 of PSDs for PM in combined sewer overflow and run-off samples for different rainfall events

		D10 (μm)				D50 (μm)				D90 (μm)			
Type	Event date	max	min	mean	SD	max	min	mean	SD	max	min	mean	SD
CSO	25 June 2013	9.57	8.77	9.14	0.34	54.0	49.4	52.1	1.8	588	382	452	81
	05 July 2013	12.10	8.58	10.23	1.25	83.9	47.1	61.7	12.4	1,160	567	746	228
	04 August 2013	10.50	8.51	9.44	0.83	65.9	49.0	55.5	6.4	960	382	663	282
	25 August 2013	10.90	8.92	9.90	0.76	67.5	56.0	60.7	4.6	722	472	579	91
	13 September 2013	21.40	12.60	17.97	2.71	91.3	57.3	83.2	11.1	672	301	630	112
	21 September 2013	15.00	10.00	12.60	1.88	106.0	55.4	68.4	21.2	872	312	487	226
	Total CSO events	21.40	8.51	11.54	3.41	106.0	47.1	63.6	13.6	1,160	301	593	193
Run-off	25 August 2013	11.30	6.28	8.89	1.69	45.8	26.4	36.8	5.4	1,060	272	397	252
	13 September 2013	9.98	5.76	7.76	1.41	43.6	25.2	33.4	6.4	986	256	363	235
	Total run-off events	11.30	5.76	8.33	1.62	45.8	25.2	35.1	6.0	1,060	256	380	237

Furthermore, the relationship between the water level of suction and total water volume in the drainage system was estimated using a hydraulic model (Infoworks CS, Innovyze, CO, USA). The difference in volume in the system between the operational water level of the interception pumps (dry weather) and the operational water level of the stormwater lifting pumps (wet weather) was taken as the in-line storage volume. Using the hydraulic model, we calculated that around 4,000 m^3 in-line storage volume exists at the Anshan experimental site.

In-line storage volume, which increases the residence time of sewage and stormwater (under light rain and initial part of heavy rain), is a main characteristic of the pump-lift drainage system. Given this large storage volume, the influence of CSO on water quality in rainfall events was delayed and weakened. In addition, the relatively high residence time of sewage was conducive to PM aggregation within the sewer system, resulting in large *in situ* particles.

3.4. PSD variation over time in CSO samples for two rainfall events

Two rainfall events on 25 August 2013 and 13 September 2013 were selected to discuss variation in PSD of PM in CSO samples over time. Most sediment accumulation in both surface and sewer waters occurs during dry periods. Hence, the number of antecedent dry days was one of the factors that influenced particle matter distribution in CSO samples. The number of antecedent dry days for these two rainfall events was 21 and 18 d, respectively (Table 1). Hence, we assumed that the accumulation of sediment for both overflow events was the same. Fig. 5 shows the variation in PSDs in samples on both dates over time, once the stormwater lifting pumps were switched on ($t = 0$).

The discharge processes observed for these two CSO events were different (Fig. 5(a) and (b)). On 25 August 2013, two stormwater lifting pumps were switched on during the initial phase of the CSO. The initial flow rate was about 5.5 m^3/s for the first 45 min. The overflow rated was reduced to 2.3 m^3/s for the last 40 min.

A relatively large variation was observed for particles in the size ranges from 10 to 100 μm, and 300 to 3,080 μm. For particles 10–100 μm in size, a monotonic increase with time was observed. As a consequence of the high flow rate caused by the pumps being switched on, a large proportion of particles between 300 and 3,500 μm in size appeared during the initial phase of the CSO. The proportion of these larger particles gradually reduced with time. The reduced flow rates in the final phase of the CSO, resulted in a decreased flush intensity during this phase. The reduction in large particles may reflect the fact that the more easily eroded sediments were flushed during the initial phase of the CSO.

On 13 September 2013, high rainfall intensity during the initial phase of this rainfall caused the water level of the system to increase rapidly. Hence, four stormwater lifting pumps were switched on within the initial 30 min. The peak flow rate was 10.2 m^3/s, lasting for more than 35 min. Thus, the flush intensity was much stronger than for the event on 25 August 2013. In general, variation in PSDs for particles 10–100 and 300–3,500 μm in size were similar to those for the event on 25 August 2013, although a greater magnitude to this variation was observed in this event. In addition, the stormwater lifting pumps were switched on again during the posterior phase of this rainfall

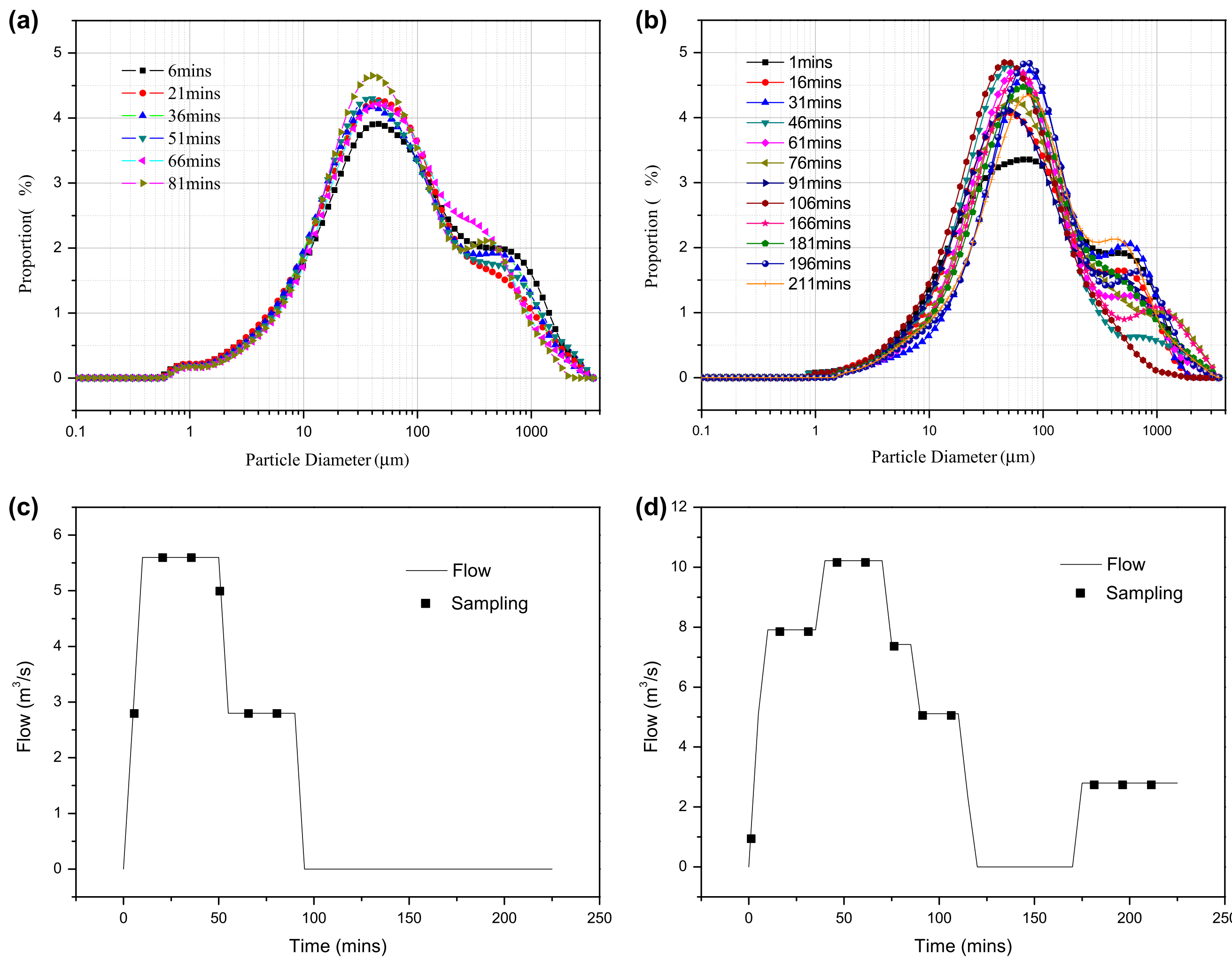

Fig. 5. Variation in PSDs with time for two rainfall events. (a and b) PSDs for CSO samples for each rainfall event (samples shown were taken at various times after the CSO discharged; their collection time is shown in min); (c and d) Flow rates for stormwater lifting pumps over time for the same rainfall events.

event. Although the flow rate increased from 0 to 2.8 m^3/s at this time, there was no significant increase in the number of large particles observed in the PSDs of samples associated with this period, suggesting that most of the easily eroded sediment had been flushed during the initial 130 min of the CSO.

3.5. PM first flush

Using a traditional assessment method, a moderated first flush of TSS was observed for all CSO events. Values for first flush strength were close to previous studies [10,11]. First flush ratios for different particle sizes were calculated using the PSDs and TSS values for each grab sample. However, this method has a drawback for small particle sizes. Values for TSS were determined by filtration with a 0.45 μm membrane [19], so particles with sizes less than 0.45 μm would not be accounted for TSS values. However, the results of PSD analysis for all CSO samples indicated that the particles with sizes less than 0.45 μm accounted for less than 0.02% of the total particles. We conclude that discounting this small fraction of particles does not seriously affect our results.

Particle matter first flush (PMFF) defined as the normalized mass of particles divided by the normalized volume fraction at any point of the normalized CSO diagram, was used to quantitatively evaluate the strength of first flush. The definition used in this study is similar to the MFFR used in previous studies [1,16–18]. Fig. 6 shows PMFF ratios (PMFF20 and PMFF30) for particles in different size ranges; a box

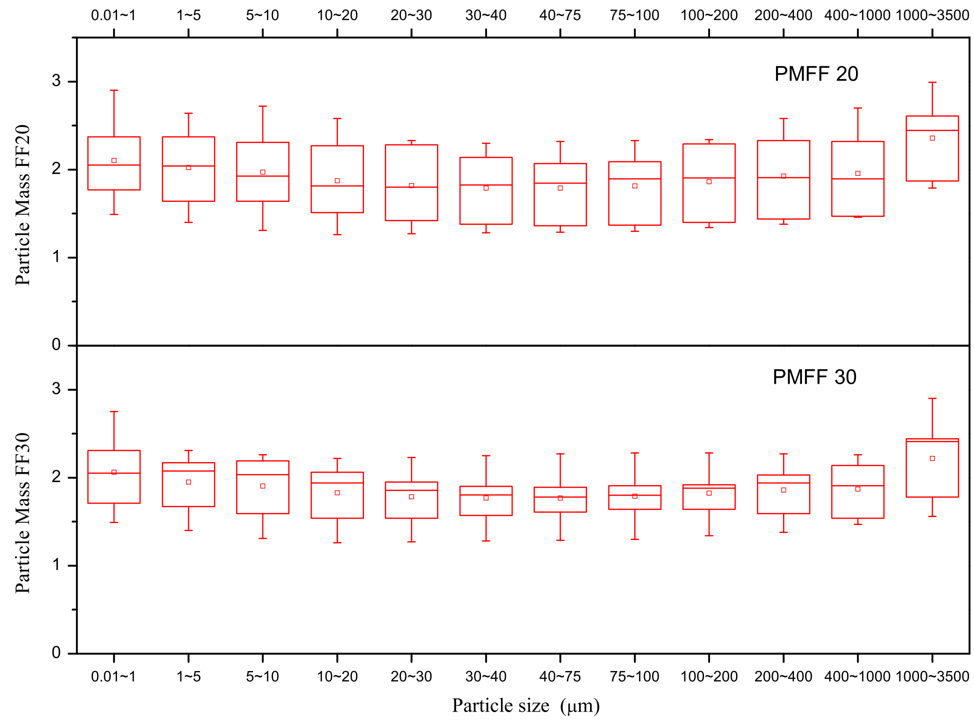

Fig. 6. Box charts for different particle size first flush ratios.

defines the ± 25% limits of the particle population, and the horizontal line marks the median. Whiskers represent maximum and minimum values, unless there are outliers.

For particles in the size range 10–3,500 μm, the median PMFF20 and PMFF30 values increase with increasing particle diameter. For particles larger than 1,000 μm, the mean values and median values for PMFF20 and PMFF30 were higher than for any other particle size. PMFF ratios for particles larger than 1,000 μm were generally larger than the analogous MFFR for TSS.

This is related to the erosion and flush of large size particles during the initial phase of the CSO. Furthermore, the first flush of particles larger than 1,000 μm was stronger than other particles in the CSO. However, the mass of particles larger than 1,000 μm only accounted for 12.9% of the total particles mass.

It should be noted that the mean values of PMFF for particles smaller than 5 μm were higher than for particles in the size range from 10 to 1,000 μm. However, the mass of particles smaller than 10 μm accounted for only a small percentage of the total particles mass in the CSO samples, with the maximum and the mean values of 6.9 and 4.1%, respectively.

More than 80% of particles were between 10 and 1,000 μm in size, constituting the main target for CSO treatment. However, the first flush strength of these particles was weaker than the MFFR for TSS. Despite this fact, the first flush ratio of particles with this size range should be considered in designing CSO treatment practices, since the performance of CSO treatment practices designed only on the TSS first flush may not be as good as expected.

4. Conclusions

This study examined the PSDs of PM in a combined sewer overflow from a high intensity urban catchment located in Shanghai, China. Our results indicated that most of the COD and TP in the CSO were within the particulate-band, while the particulate-band TN only accounted for 50% of TN in the CSO. A clear effect of sample storage time on PSDs of PM was observed, thus we recommended that PSDs of PM in CSO, DWF and run-off samples should be analyzed within 6 h of collection.

Analysis of the PSDs of PM in CSO samples from the high intensity urban catchment in Shanghai, China, showed that the particles ranged in size from 0.41 to 3,080 μm. The mean D50 and D90 values for PM in both CSO and run-off samples were higher than for previous studies. The high flow rates caused by stormwater lifting pumps contributed to the larger particle sizes recorded in this study.

Based on the PMFF defined in this study, the strength of the first flush for different particle sizes was quantitatively evaluated. The first flush strength for particles in the size range 10–1,000 μm was weaker than the MFFR for TSS. More than 80% of the particles in the CSO ranged in size from 10 to 1,000 μm, suggesting this fraction should be the main target for CSO treatment. Not only the TSS first flush, but also the first flush of particles within the range from 10 to 1,000 μm should be considered in designing CSO treatment practices.

Knowledge of the PSD and its variation over time, and between events, is critical for CSO treatment designs and optimization of wastewater treatment plants in Shanghai and other cities. Given the prevalence of combined sewer systems in China, our PSD data provide an important input for modeling treatment unit operations. Our results suggest that determining the COD, TP, and TN for different particle sizes is important for future research.

Acknowledgments

This study was financially supported by National water pollution control and management technology major projects (2011ZX07303-002). We also acknowledge the technical support of other group members.

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