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RESEARCH ARTICLE

Temporal variation of heavy metal pollution in urban stormwater runoff

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Abstract Stormwater runoff from three types of urban surfaces, a parking lot, a street, and a building roof, was monitored during four rainfall events that occurred in the one-year period from June 2009 to June 2010. The event mean concentrations (EMC) of dissolved copper (Cu), lead (Pb), zinc (Zn), manganese (Mn), and iron (Fe) exceeded China's National Water Quality Standards for Surface Water. The degree of heavy metal contamination was related to the type of underlying surfaces. Additionally, the concentration of dissolved heavy metals peaked shortly after the runoff began and then declined sharply as a result of adequate flushing. First flush effects of varying degrees were also observed during all of the monitored rainfall events based on the first flush ratio (FF_{25}). Redundancy analysis revealed that four environmental variables (rainfall depth, intensity, antecedent dry weather period and type of underlying surface) had significant effects on the strengths of the first flush effects, accounting for 72.9% of the variation in the FF_{25}. Dissolved metals presented varying first flush effects on different underlying surfaces that occurred in the following relative order: parking lot > roof > road for low intensity and high runoff volume rainfall events; parking lot > road > roof for high intensity and low runoff volume events. The relative strength of the first flush for dissolved heavy metals was Fe, Mn > Cu, Zn > Pb.

Keywords urban stormwater, heavy metal pollution, temporal variation, event mean concentration, first flush effect, redundancy analysis

1 Introduction

Urbanization has dramatically increased the amount of impervious surfaces, which has contributed to flooding and runoff contamination; accordingly, urbanization poses challenges to storm water management. In China, in addition to the impact of rapid urbanization and industrialization, direct discharge of stormwater runoff into surface water bodies contributes to the degradation of stream water quality [1,2]. During rainwater events, rain washes dust out of the atmosphere and from impervious urban surfaces, after which, dissolved, colloidal and solid constituents in a heterogeneous mixture composed of organic and inorganic compounds, nutrients, oils, greases and heavy metals are carried to local water bodies via stormwater runoff [3,4]. Previous studies have reported that several water quality variables including nutrient elements (nitrogen and phosphorus) and suspended solids, in stormwater runoff exceed the standards of surface water quality [5–7]. In addition, stormwater runoff has been found to contain high levels of heavy metals such as Pb, Zn, and Cd as a result of runoff from roads and roofs [8,9]. Heavy metals in stormwater runoff are present in dissolved (< 0.45 μm) and particulate (> 0.45 μm) forms, and those in particulate form can be transformed to the dissolved form under certain conditions [10]. Owing to the bioavailability of dissolved heavy metals, their concentrations have been closely linked to the toxicity of stormwater runoff [11].

The first flush refers to the delivery of a disproportionately large load of constituents during the early part of the runoff hydrograph. Whether this effect exists or not and its strength strongly influence the type and size of storm water management facilities that are required for an area. Therefore, in this study, samples of roof, road and parking lot runoff from four rainfall events that occurred during the period from June 2009 to June 2010 were collected and analyzed. The specific objectives of this study were: 1) to evaluate the level of contamination with dissolved heavy metals in roof, road and parking lot runoff; 2) to investigate the temporal effects of heavy metal pollution in stormwater runoff from different underlying surfaces; 3) to identify key parameters influencing the strength of first flush

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effects. The findings of this study are expected to facilitate the development of effective strategies for control of metal elements in urban runoff.

2 Materials and methods

2.1 Experimental site

The study was conducted in Taoranting Park and its surrounding area, which is a mixed commercial and residential area adjacent to the South Moat in downtown Beijing from June 2009 to June 2010. Taoranting Lake and the South Moat are two of the three key surface water bodies approved by the Beijing Water Authority as part of a water quality improvement project in 2009. This region has a temperate monsoon climate, with an annual mean temperature of 12.4°C and an annual mean rainfall of approximately 600 mm. About 74% of the rainfall occurs in summer (June to August). The drainage area of the study site is 1.01 km^2, with impervious underlying surface accounting for 48.6% (including roads, sidewalks, parking lots, etc.). Three sampling sites representing the main types of land use in urban regions were selected.

- Site 1: roof covered with tar paper;
- Site 2: Caishikou Street, which has a high traffic volume;
- Site 3: a parking lot in front of the north gate of Taoranting Park.

2.2 Sampling and analysis

2.2.1 Sampling and laboratory analysis

Samples of runoff were collected from the three sites described above. All water samples were collected into 600 mL plastic bottles, with the collection period beginning at the generation of runoff and ending when the flow receded to the dry weather water level. Samples were collected at 2-min intervals during the first 10 min, followed by 5-min intervals in the next 20 min, 10-min intervals in the next 30 min, and then 15-min intervals until the runoff ceased. The collected runoff and rainfall samples were transported to the laboratory within one hour. Rainfall intensity data were recorded using an automated weather station installed in the core of the study area.

The pH of all samples was measured immediately upon arrival in the laboratory and then stored at 4°C until subsequent analysis. Samples were filtered through 0.45 μm mixed fiber microporous membranes and the filtrates were then tested for the presence of dissolved metals (Mn, Pb, Zn, Cu, Fe) by inductively coupled plasma (ICP) atomic emission spectrometry [12]. Owing to equipment failure, values for Fe in Event #1 and Pb in Event #4 were not determined. A total of 259 samples were collected and analyzed.

2.2.2 Event mean concentration (EMC)

Even though the concentration of a pollutant often varies by several times during a rainfall event, a single index known as the event mean concentrations (EMC) can be used to characterize runoff constituents [13]. The EMC represents a flow-weighted average concentration computed as the total pollutant mass divided by the total runoff volume using the following Equation [14]:

$$EMC = \frac{M}{V} = \frac{\int_0^t C_t Q_t \mathrm{d}t}{\int_0^t Q_t \mathrm{d}t} \cong \frac{\sum C_t Q_t \Delta t}{\sum Q_t \Delta t}, \tag{1}$$

where, M is the total mass of pollutants over the entire event duration (g); V is the total volume of flow over the entire event duration (m^3); t represents time (min); C_t is the concentration of a pollutant ($\mathrm{mg \cdot L^{-1}}$); Q_t is the variable flow ($\mathrm{m^3 \cdot min^{-1}}$); and Δt is a discrete time interval (min).

2.2.3 First flush effect

The first flush is defined as the initial period of stormwater runoff during which the concentrations of pollutants are substantially higher than those observed during latter stages of the storm event. Analysis of the first flush effect in this study was based on the relationship between the cumulative mass curve and the cumulative runoff volume curve. The 45° line ($y = x$) represents the case in which pollutant loadings in runoff are constant throughout the storm event. The percentage deviation of the curve from the diagonal ($y = x$) then serves as a reference for the strength of the first flush [15]. The strength of the first flush can be quantified for each storm event and for each water quality parameter using a mass first flush ratio, MFF. FF_{25} is the total pollutant load transported by the first 25% of the runoff. The following equation was used to quantify the strength of the first flush [16]:

$$FF_{25} = \frac{M_{(t)}}{V_{(t)25}}, \tag{2}$$

where $M_{(t)}$ is the dimensionless normalized cumulative load of heavy metals and $V_{(t)25}$ is the dimensionless normalized cumulative volume corresponding to 25% of the total runoff volume. For example, an FF_{25} equal to 2 indicates that 50% of the pollutant mass is contained in the first 25% of the runoff volume.

2.3 Characteristics of rainfall events

The record of the automated weather station installed at the study area revealed that there were nine rainfall events in which the rainfall exceeded 3 mm from June 2009 to June 2010. Among them, only seven generated runoff. Due to personnel safety concerns, samples were not collected under thunderstorm conditions; therefore, four effective rainfall events were monitored.

As shown in Table 1, the characteristics of the monitored rainfall events were diverse. The maximum event mean intensity, 26.0 mm·h^{-1}, occurred during Event #4 and the minimum, 2.5 mm·h^{-1}, occurred during Event #1. According to the definition of rainfall intensity defined by the National Meteorological Department, the four rainfall events monitored in this study were good representations of high, medium and low intensity rainfall events typically found in the Beijing region.

3 Results and discussion

3.1 Water quality of stormwater runoff

Descriptive statistics, including the mean, maximum and minimum of the monitored water quality parameters for each type of runoff, are shown in Table 2. Zn concentrations ranged from 0.048 mg·L^{-1} to 2.088 mg·L^{-1} in the runoff from the three types of surfaces. The maximum concentration of dissolved Zn (2.088 mg·L^{-1}) in the roof runoff exceeded both the China National Environmental Quality Standards for Surface Water (GB3838-2002) and the Discharge Standard of Water Pollutants in Beijing (DB11/307-2005). Regarding the China National Environmental Quality Standards for Surface Water (GB3838-2002), the maximum concentrations of dissolved Pb in these three types of runoff all exceeded the standards. For Mn and Fe, the maximum concentrations in the road runoff were 0.200 mg·L^{-1} and 0.425 mg·L^{-1}, respectively. For the parking lot, these numbers were 0.208 mg·L^{-1} and 0.872 mg·L^{-1}, respectively. For both of these surfaces, the values for Mn and Fe were 1 to 3 times higher than the Water Quality Standard for Surface Water Source of Centralized Drinking Water. Previous studies have demonstrated that particle adsorbed heavy metals played a dominant role in stormwater runoff [17,18]. The results of the water quality analysis conducted herein demonstrated that dissolved heavy metals can not be ignored and also underlined the threat of direct discharge of stormwater runoff into natural waters.

3.2 Spatial variation of EMC

The water quality of stormwater runoff is affected by the processes of pollutant buildup and washoff, which are closely related to the type of underlying surface. In this study, the water quality of runoff from different underlying surfaces is discussed. As shown in Table 2, the EMC values of different surface types were in the order of: road > parking lot > roof. The EMCs of road runoff were higher than those for the other two surfaces, which was mainly due to the complexity of the activities in the study region. A host of factors affected the pollutant buildup on the traffic artery, including traffic flow and the presence of a commercial district that includes shopping centers and restaurants next to the road. Roof and road runoff were the major pollution sources of dissolved Zn. Galvanized roofing materials and downpipes caused the high concentration of dissolved Zn in the roof runoff, while tire wear and lubricating oil were the main sources of dissolved Zn in the road runoff [14,21]. These results suggest that there is an environmental benefit to using fewer Zn compounds and fewer galvanized materials. High levels of dissolved

Table 1 Characteristics of four rainfall events

event	date	rainfall depth /mm	duration /h	mean intensity /(mm·h^{-1})	maximum intensity /(mm·h^{-1})	antecedent dry weather period/d
1	2009–06–18	4.2	1.67	2.5	28.8	10
2	2009–07–17	46.6	5.17	9.0	28.8	4
3	2009–08–01	21.8	2.58	8.5	57.6	2
4	2010–06–01	22.6	0.87	26.0	252.0	1

Table 2 Pollutant concentration ranges for each type of runoff/(mg·L^{-1})

	roof runoff			road runoff			parking lot runoff		
	min	max	mean	min	max	mean	min	max	mean
Cu	0.003	0.019	0.008	0.004	0.021	0.011	0.005	0.013	0.008
Mn	0.010	0.115$^{a)}$	0.039	0.031	0.200$^{a)}$	0.108$^{a)}$	0.017	0.208$^{a)}$	0.103$^{a)}$
Pb	0.019	0.054$^{a)}$	0.031	0.022	0.050$^{a)}$	0.034	0.014	0.055$^{a)}$	0.035
Zn	0.170	2.088$^{a,b)}$	0.778	0.055	0.924	0.315	0.048	0.146	0.089
Fe	0.027	0.151	0.073	0.090	0.425$^{a)}$	0.256	0.109	0.872$^{a)}$	0.365$^{a)}$

Notes: a) the data exceeded China National Environmental Quality Standards for Surface Water (GB3838-2002) [19]; b) the data exceeded Discharge Standard of Water Pollutants in Beijing (DB11/ 307-2005) [20]

Pb, Mn, and Fe were detected in the road and parking lot runoff. Diffusion of exhaust gases generated from the combustion of petrol and diesel fuels was the dominant factor influencing Pb pollution. Despite attempts by China to implement the use of unleaded petrol since 2000, the petrol in use still contains lead at a concentration of 0.013 $g\cdot L^{-1}$. The heavy traffic in the metropolitan area has also aggravated pollution. Ferromanganese has been widely used in vehicle, road and building materials, which is the main reason for the high levels of Fe and Mn in road and parking lot runoff. As shown in Fig. 1, the spatial distribution of runoff pollution of Fe and Mn is similar. This phenomenon can be explained by the results of a previous study conducted to investigate the chemical form of heavy metal elements in freeway runoff [22] in which Fe-Mn oxide was found to be 66.3% elemental Mn. In addition, the Fe-Mn oxide band is in a form that can be effectively leached under natural rainy conditions.

3.3 Temporal distribution of runoff pollution

3.3.1 Variation in dissolved heavy metal concentrations in runoff during rainfall events

Figure 2 presents the variation in dissolved heavy metal concentrations in roof, road and parking lot runoff during rainfall Events #2 and #4. These rainfall events differed, in that Event #2 was bimodal and Event #4 was a single peak event. In Event #2, a secondary flush effect phenomenon common to bimodal rainfall events occurred during the heavy metal leaching process. Event #4 was a typical single peak rainfall event with a short duration and high intensity. During this event, the concentrations of dissolved heavy metals peaked shortly after the runoff was generated. Because of adequate flushing, the concentrations declined to 1/3 to 1/7 of the peak value approximately 30 min after the runoff was generated.

The concentrations of dissolved metals were directly associated with the rainfall intensity. As shown in Fig. 2, the concentrations of the metals peaked slightly after the rainfall intensity peaked. After the initial stage, the concentrations of dissolved metals decreased sharply. This regular pattern is consistent with the results of previous studies [23,24]. However, the characteristics of the roof runoff were slightly different. Concurrent with the decrease in precipitation and rainfall intensity, the concentrations of dissolved metals rose slightly. One potential reason for this was that the flushing effect played an important role when the rainfall intensity was high, but in the later period the ponded water on the roof was only slightly disturbed owing to the low rainfall intensity. Heavy metals are also more likely to leach out with longer

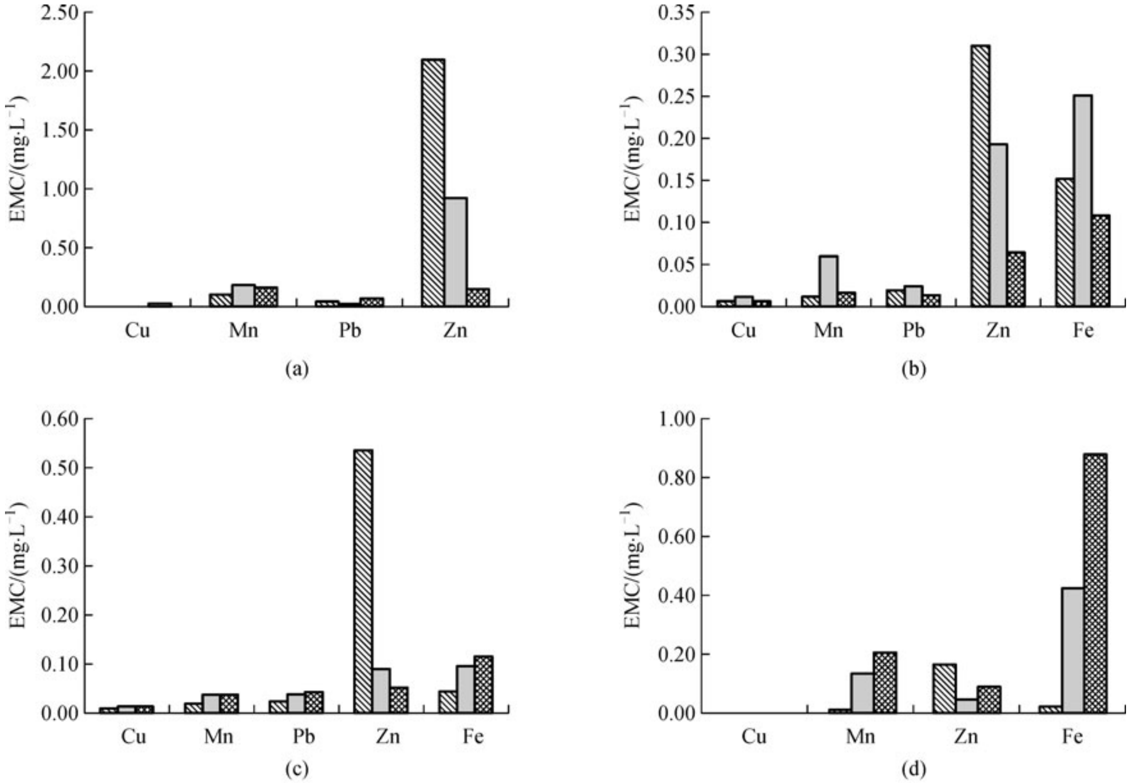
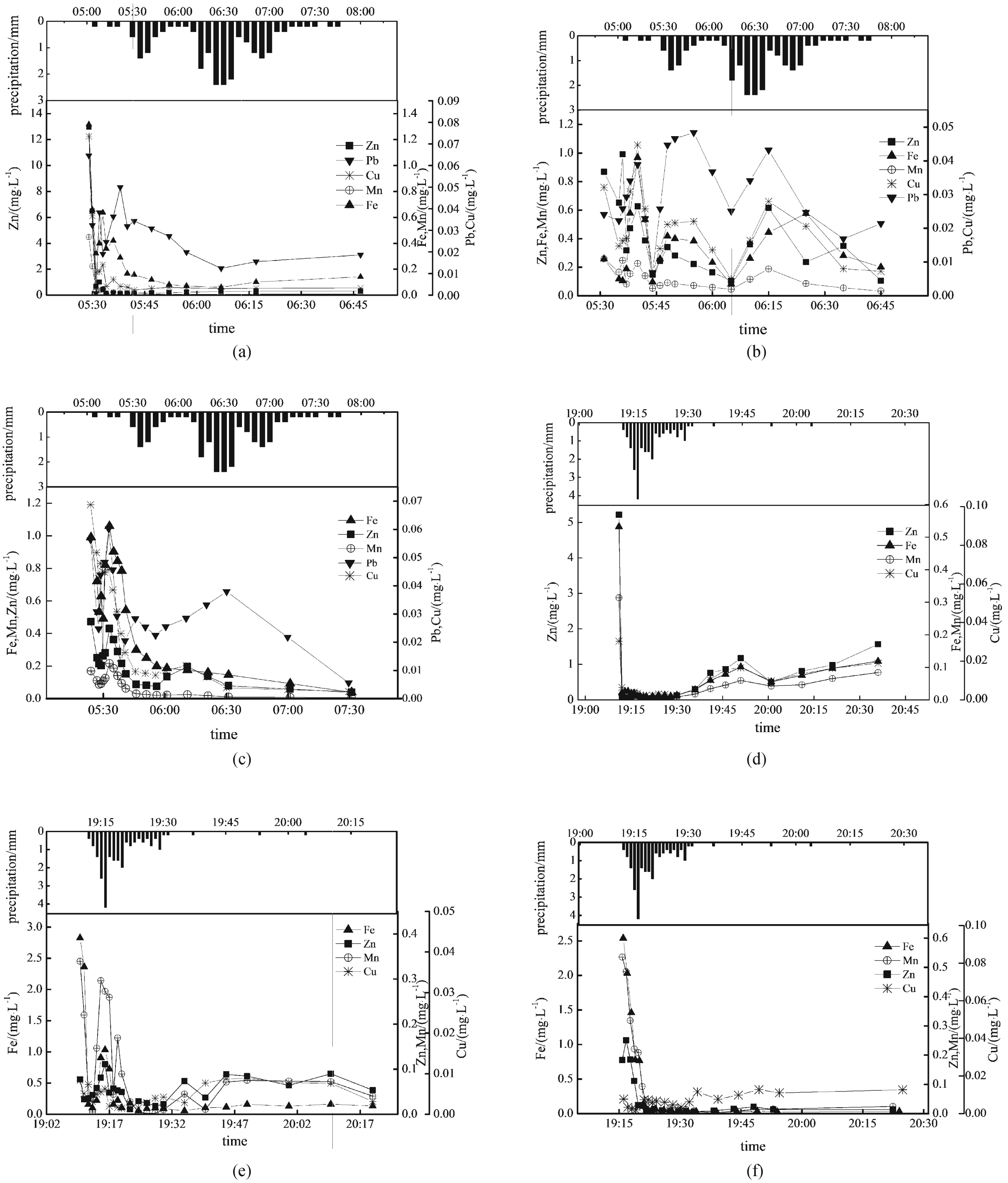


Fig. 1 Quality of stormwater runoff from different underlying urban surfaces: rainfall events 2009–06–18 (a), 2009–07–17 (b), 2009–08–01 (c), and 2010–06–01 (d)



(a) (b) (c) (d) (e) (f)

Fig. 2 Temporal variation of pollutant concentrations during the rainfall events: rainfall event 2009–07–17, roof (a), road (b), and parking lot (c); rainfall event 2010–06–01, roof (d), road (e), and parking lot (f) hydraulic retention times. Additionally, pH may have influenced heavy metals leaching. Specifically, the average pH values for roof, road and parking lot runoff over these four rainfall events were 6.2, 7.0 and 7.6, respectively. The acidic environment of the roof runoff was conducive to the leaching of heavy metals. As shown in Fig. 2, the concentrations of dissolved heavy metals in the road runoff fluctuated violently during storm events. The deposition and accumulation of heavy metals on the road resulted from traffic activities, vehicular component wear, fluid leakage, pavement degradation, and roadway maintenance [25,26]. Vehicles still traveled on rainy days, so the pollution occurred continuously. All of these factors resulted in the discharge pattern of the road runoff being more complex than that from the other areas.

3.3.2 First flush effect

The first flush effect was investigated by plotting the cumulative mass curve against the cumulative runoff volume curve. The M(V) curves for dissolved metals are shown in Fig. 3 for Event #2 and Event #4. Event #2 was a low intensity, high runoff volume rainfall event and Event #4 was a high intensity and low runoff volume event. Most of the curves were above the diagonal line, illustrating the existence of a first flush phenomenon. There was an interesting phenomenon, which is different from conclusions of previous studies [6,27], in which rainfall events of high intensity had a strong first flush effect of total heavy metals. In this study, the strength of the first flush effect on dissolved heavy metals in Event #2 was a bit stronger than that of Event #4.

To quantify the strength of the first flush effect, the FF_{25} values of the four rainfall events were calculated (Table 3). Dissolved metals showed varying strengths of first flush effects on different underlying surfaces that occurred in the following order: parking lot > roof > road for low intensity and high runoff volume rainfall events and parking lot > road > roof for high intensity and low runoff volume events. The sources of pollution on the road and parking lot were similar, but the strength of the first flush effects on the road and parking lot differed. The potential cause for this phenomenon may be that, unlike the situation for other underlying surfaces, the pollution occurred continuously on road surface during the rain. The box chart in Fig. 4 shows that the relative strengths of the first flush effects for dissolved heavy metals were Fe, Mn > Cu, Zn > Pb. These results are in accordance with those of previous studies [27,28]. In a study concerning morphological characteristics of heavy metals in soil media along a road [22], the relative soil retention capacity of heavy metals was Pb > Cu > Mn. Obviously, a heavy metal for which the retention capacity with soil particles is strong is not easily leached out in dissolved form during a rainfall event, so the strength of the first flush effect on such metals would be low. Overall, these results suggest that factors such as site and characteristics of rainfall events resulted in variations in the first flush effects. As a result of the numerous potential impact factors and their randomness, the results of previous studies concerning the relationships between the strength of a first flush effect and its influencing factors vary [27–29].

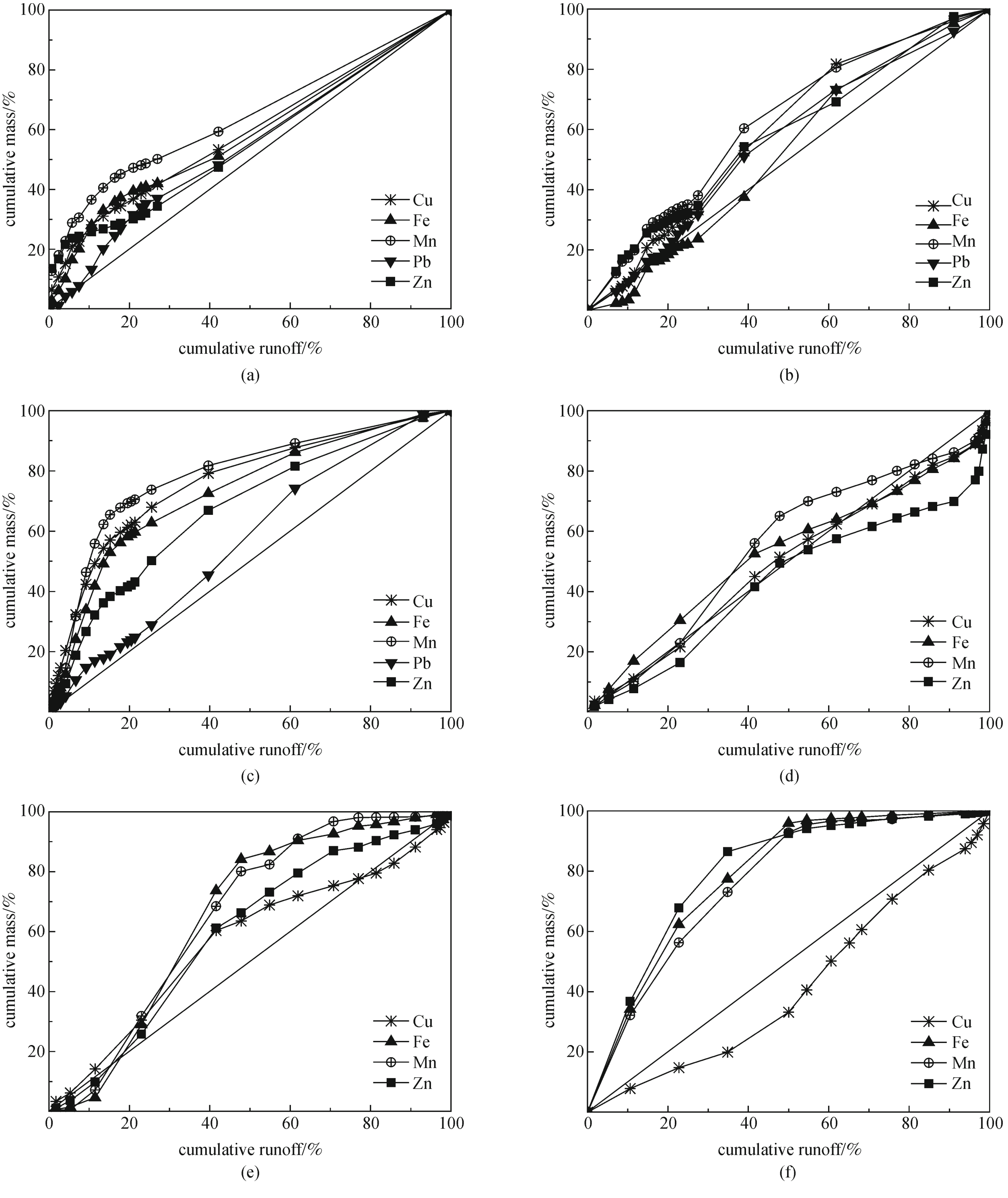
To quantify these influencing factors, Statistical Product and Service Solutions (SPSS) 18.0 was used to compute the Pearson correlation coefficients between FF_{25} and the potential impact factors. No correlation was observed, but the results of redundancy analysis showed that rainfall depth, intensity, antecedent dry weather period and the type of underlying surface had significant effects on the strength of the first flush effect, accounting for 72.9% of the variation in the first flush ratio FF_{25}.

4 Conclusions

In rainfall Events #1, #2, #3, and #4, the EMCs of dissolved Pb, Zn, Mn, and Fe exceeded China’s National Water Quality Standards for Surface Water to varying degrees. These findings suggest that the dissolved heavy metals should not be ignored and underline the threat of direct discharge of stormwater runoff into natural waters. The accumulation of heavy metals varied among surfaces. Roof and road runoff were the main pollutant sources of dissolved Zn, while high levels of dissolved Pb, Mn, and Fe were most likely to be detected in road and parking lot runoff.

In this study, temporal effects of heavy metal pollution in stormwater runoff were investigated. The concentration of dissolved heavy metals peaked shortly after the runoff was generated. Through adequate flushing, the concentrations declined to 1/3 to 1/7 of the peak values. During the monitored rainfall events, dissolved metals presented varying first flush effect strengths on different underlying surfaces. The relative order was parking lot > roof > road for low intensity and high runoff volume rainfall events and parking lot > road > roof for high intensity and low runoff volume events. Due to the spatial variation of first flush effects in an urban area, different interception ratios of initial rainfall-runoff volumes should be considered in different catchments for runoff pollution control. The relative strength of the first flush effects for dissolved heavy metals was Fe, Mn > Cu, Zn > Pb. For the sake of simplification, prior controlled pollutants can be selected as representative or control indicators in runoff pollution monitoring and management.

Redundancy analysis demonstrated that the characteristics of sampling sites and rainfall events had significant effects on the strength of the first flush effect. Four environmental variables (rainfall depth, intensity, antecedent dry weather period and the type of underlying surface) accounted for 72.9% of the variation in the first flush ratio FF_{25}. Given that the monitoring data in this



(a) (b) (c) (d) (e) (f)

Fig. 3 First flush effects of stormwater runoff pollution in different underlying urban surfaces: rainfall event 2009–07–17, roof (a), road (b), and parking lot (c); rainfall event 2010–06–01, roof (d), road (e), and parking lot (f)

Table 3 Index values of first flush effect strength (FF_{25}) for four rainfall events

	2009–06–18			2009–07–17			2009–08–01			2010–06–01		
	roof	road	parking lot	roof	road	parking lot	roof	road	parking lot	roof	road	parking lot
Cu	1.78	1.49	0.98	1.63	1.35	2.66	3.07	1.48	1.92	1.06	1.36	0.64
Mn	2.01	1.20	1.15	1.99	1.55	2.91	2.45	1.36	1.68	1.31	1.66	2.36
Pb	1.01	0.91	0.80	1.38	1.18	1.24	0.39	1.33	1.09	–a)	–	–
Zn	2.41	1.47	1.32	1.35	1.39	1.97	1.77	1.80	1.78	0.90	1.42	2.62
Fe	–	–	–	1.67	0.98	2.47	1.55	2.06	2.24	1.22	1.68	2.50

Note: a) no data

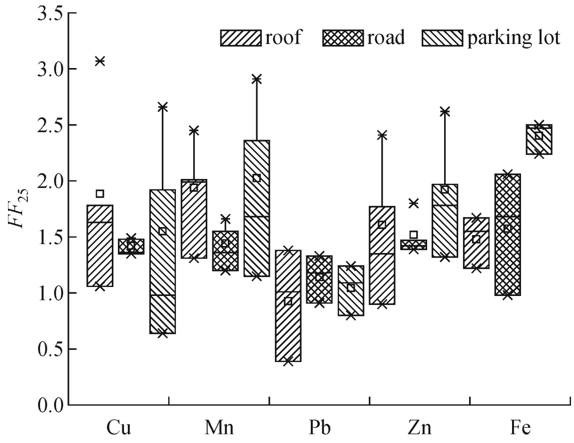


Fig. 4 Box distribution charts for FF_{25}

study is limited, more research is required to clarify this relationship.

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