



Distributions and transport of typical contaminants in different urban stormwater runoff under the effect of drainage systems

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ABSTRACT

In recent years, the pollution of receiving water induced by urban stormwater runoff has gained considerable attention. Meanwhile, rainwater is also a potential and valuable water resource, which may effectively mitigate the increasing water shortage. In the present study, the distributions and transport of nutrients and heavy metals in different stormwater runoff of two residential communities (site A and B) under the effect of drainage systems were discussed in detail. Site A with combined drainage system was located in old district featured with a lot of big manufacturing and chemical factories in suburb. Differently, site B with separate system was situated in new district occupied by small shops and residential quarters. The results indicated that air quality was a significant influence factor and reference index in the decision-making of rainwater harvesting and reuse. For nutrients, the anthropic activities contributed adequately ammonia nitrogen and total phosphorus to the road runoff (RD) and sewer overflows, especially in site A with combined drainage system. However, the roof runoff (RF) presented the highest event mean concentrations (EMC) of nitrate and nitrite nitrogen, probably due to the high runoff coefficient of roof and low interference from human activities on dry deposition. Zn(II), Cu(II), and Pb(II) concentrations showed a similar variation pattern, although there were some small differences owing to the erosion of metal materials from rooftops and street lamps. Compared with the "Standards for Drinking Water Quality" in China, the direct rainfall and RF in site B could be harvested and utilized for potential drinking water supply from the perspective of the studied parameters. For site A, at least 33.8% of NH₄⁺-N and 50.8% of NO₃⁻-N in RF must be removed before reuse. The results obtained in this research would provide an important assistance and support in urban rainwater reuse as well as runoff pollution control.

Keywords: Pollution distribution; Event mean concentrations; Surface runoff; Combined drainage system; Separate drainage system; Rainwater harvesting; Water quality

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1. Introduction

In recent years, due to the rapid urbanization and ever-increasing population, the atmosphere and ground surface such as roof, road in urban areas are withstanding severe pollution. In wet weather, a great deal of pollutants are thereby swept up and discharged into the receiving water accompanied with wet deposition and stormwater runoff. It is widely reported that the urban runoff has been one of the main reasons which cause significant damage to the aquatic environment [1-3]. Therefore, characterizing the water quality of different surface runoff has been an essential premise for subsequent management measures. Moreover, in order to better protect the urban water environment, it is greatly needed to understand the distributions and transport of typical contaminants in different kinds of stormwater runoff.

As is known to all, urban drainage systems usually consist of two main forms: combined one and separate one. After different surface runoff is discharged into the sewers and mixed with wastewater as well as sewer deposits, a lot of complicated processes including physical, chemical, and biological may take place, especially in combined sewers [4-6]. Therefore, the water quality of the end effluent of sewers, which has more direct connections with receiving water, should also be evaluated. So far, the water quality of different surface runoff such as roof runoff (RF) and road runoff (RD) in wet weather has been extensively analyzed in many studies [4–11]. However, only a few of them have further evaluated the stormwater runoff after in-sewer processes [4-6]. Furthermore, these few researches most focused on combined sewers [4-6] while the comparable research under separate drainage system is rarely reported. Considering that the difference in drainage systems would inevitably bring on the diversity in pollution distributions and transport, a comparison of typical contaminant concentrations under different drainage systems is in great need.

Besides its pollution, in many countries and regions, especially the arid and semiarid ones, the rainwater is often harvested, stored, and treated as a promising water resource [12,13]. Considering the storage time and treatment costs, it would be more cost-effective and convenient to collect slightly polluted runoff than badly polluted one. Nitrogen (such as NH_4^+ -N, NO_2^- -N, and NO_3^- -N) and phosphorus are the common nutrients in surface runoff [7,14], which would lead to the deterioration or even eutrophication of water body [15] and thereby shorten the storage period of rainwater. Meanwhile, heavy metals such as Zn(II), Cu(II), and Pb(II) are also critical factors, which

would pose potential toxicity to the aquatic organisms [16,17] and influence the safety of rainwater reuse. Therefore, understanding the distributions and transport of nitrogen, phosphorus, and heavy metals would provide important information for rainwater harvesting, storage, and reuse.

The purpose of this study was to investigate the distributions and transport of nitrogen, phosphorus, and heavy metals in different urban stormwater runoff. The water quality of end effluents from combined sewers and separate ones were also characterized and compared to reveal the effect of drainage systems. Besides, the feasibility of rainwater utilization from different surfaces of two sampling sites was further evaluated. The results obtained in this study would provide significant reference on runoff pollution control and rainwater harvesting.

2. Experimental methods

2.1. Sampling sites and methods

Two residential communities were selected as sampling sites in a big seaside city of southeast China. Site A had been built for about 30 years with combined drainage system in old district while site B was constructed within the last decade with separate system in new district. The old district was featured with more than 100 big manufacturing and chemical factories in suburb, and innumerable small shops such as auto servicing, supermarkets, and laundries spreading around the residential quarters. Besides, a busy outdoor vegetable market located near site A. By contrary, the new district was more occupied by small shops and residential quarters, and few manufacturing or chemical factories were permitted to establish. Only a small car-washing shop was built within site B. Apart from the above differences between the two chosen communities, site A and B had similar total area (7–8 ha), population density (about 1,000 persons/ha), and green coverage rate (32-35%), so as to make the results comparable. In both site A and B, the green space was built as sunken one and no overflows were found in most rainfall events. Therefore, the lawn runoff was not taken into consideration in the present study.

The weather forecast was paid special attention to obtain the timely rainfall information. Three kinds of runoff including RF, RD, and sewer overflows (SO) were sampled in each community with 500-ml plastic bottles since the continuous runoff began. The RF samples were taken in the corresponding rainwater pipes while the RD samples in the selected roadside gullies and SO in the end of the sewers into the receiving water. Besides, two plastic round salvers (depth = 5 cm and diameter = 50 cm) were applied to collect the samples of direct rainfall (DR) in site A and B, respectively, in order to get background pollution status. The sampling intervals were gradually prolonged with the increase of rainfall duration, namely 5, 10, and 30 min within the runoff time of begin-30, 30–60 and 60 min-end, respectively. When the runoff ended the sampling was stopped.

2.2. Analytical methods

 NH_4^+-N , NO_2^--N , and NO_3^--N , total phosphorus (TP), Zn(II), Cu(II) and Pb(II) were measured according to Standard Methods [18]. All the concentrations in this study refer to event mean concentrations, which were calculated by the methods described in Li's studies [19]. Meanwhile, the Zn(II), Cu(II), and Pb (II) concentrations referred to the total ones including soluble and granular portions. And they were determined with a P-4010 inductively coupled plasma emission spectrometer (Hitachi Ltd. Japan) after the samples were digested by HNO₃ solution (50%, v/v).

3. Results and discussion

3.1. Rainfalls characteristics

To characterize the water quality of various stormwater runoff under different drainage systems, six rainfall events were sampled between June 2011 and May 2012. The rainfall characteristics were presented in Table 1. The EMC of different pollutants in six rainfall events were averaged and showed in the following text.

3.2. Distributions and transport of nitrogenous pollutants under different drainage systems

Fig. 1 presented the EMC of nitrogenous pollutants from DR, RF, RD, and SO in site A and B. As stated

Table 1Characteristics of the six rainfall events

above, site A was located in the old district, which was featured with a great deal of big factories while site B was in the new district with more small shops. As a result, for ammonia, nitrate, and nitrite nitrogen of DR, their EMC values in site A achieved about 2.2, 3.9, and 2.3 times as that in site B, respectively. It indicated that the background pollution for site A was much more serious than site B.

For individual ammonia nitrogen (Fig. 1a), its concentrations in RF did not exhibit an evident improvement compared to DR. Considering the roof materials themselves did not contain any nitrogen content, dry/wet deposition would be the dominating source for NH_4^+ -N in RF. It could be included that the atmospheric deposition was not a notable contributor to the ammonia nitrogen in the stormwater runoff. However, the RD showed a sharp increase in NH_4^+ -N concentration, no matter in site A or B. As the dry/ wet deposition did not supply considerable amount of ammonia nitrogen, this rise in EMC of NH_4^+ -N should be mainly attributed to anthropic activities, such as leakage of domestic waste and wastewater.

Moreover, after mixing with the wastewater and sewer deposits, the SO showed a drastic improvement in NH₄⁺-N concentration, especially for site A. It was found that the ammonia nitrogen concentrations in SO of site A and B reached 76.5 and 8.4 times as that in DR, respectively. Undoubtedly, the substantive ammonia nitrogen load mostly originated from the dry weather flow of wastewater and sewer deposits. Compared to the combined sewers in site A with sufficient wastewater, the separate sewers in site B was only mistakenly discharged into a small amount of sewage. Therefore, its improvement in NH⁺₄-N EMC was not in the same magnitude as site A. It can be concluded that the combined sewers contributed more NH₄⁺-N to the sewer overflows than storm sewers. And the effect of combined drainage system on the water quality of SO was more notable than separate one.

In Fig. 1(b) and (c), the nitrate and nitrite nitrogen presented a similar change pattern, which was quite

Date	Rainfall length (min)	Rainfall depth (mm)	Max rainfall intensity (mm/h)	Average rainfall intensity (mm/h)	Antecedent dry days (d)	
2011-07-17	335	66.8	90.0	12.0	0.7	
2011-08-24	164	8.2	24.0	3.0	0.8	
2011-10-13	520	14.5	18.0	1.7	0.3	
2012-02-22	167	14.0	42.0	5.0	5.6	
2012-03-20	63	3.6	18.0	3.4	6.5	
2012-05-09	116	11.4	54.0	5.9	4.5	



Fig. 1. Distributions of nitrogenous pollutants under different drainage systems in six rainfalls: (a) ammonia nitrogen, (b) nitrate nitrogen and (c) nitrite nitrogen.

different from ammonia nitrogen. The RF took up the highest EMC while DR, RD, and SO did not change much. The extraordinary high value of RF may be caused by two reasons: (1) there were scarcely any anthropic activities on the roof and thereby the dry deposition process would not be disturbed by passengers and/or vehicles as frequently as road; (2) the roof were covered with impervious materials with very high runoff coefficient while the road would infiltrate and retain an impressive part of runoff, especially the first flush with serious pollution [20]. Consequently, the EMC of nitrate and nitrite nitrogen in RF evidently exceeded that in RD. Moreover, their concentrations in SO of site A and B did not show a similar huge increase as ammonia nitrogen. This phenomenon was probably due to the reason that the domestic wastewater and sewer deposits did not contains as much NO_2^- -N or NO_3^- -N as NH_4^+ -N [21,22].

3.3. Distributions and transport of TP under different drainage systems

From Fig. 2, which described the TP EMC of DR and different stormwater runoff, it can be seen that DR only contained a small concentration of TP, with values of 0.16 ± 0.06 and $0.04 \pm 0.02 \text{ mg/L}$, respectively for site A and B. Due to the accumulated atmosphere deposition, the RF achieved 3.1 and 1.9 times of TP as that in DR of site A and B. The RD still acquired a higher TP value than RF, probably because the additional contribution of human activities apart from dry/wet deposition. The TP concentration in site A was 5.7 times as that in DR while the corresponding value in site B was 11.3 times. The RD of site B was clearly influenced by human activates to a greater degree in TP than site A. The reason may lie in the fact that besides the common leakage from daily lives of residents, there was a car-washing shop within site B, which generated and discharged a lot of wastewater containing phosphorus detergents to the ground and storm sewers.

In terms of TP in SO, similar to the change pattern of ammonia nitrogen, its EMC also exhibited a huge improvement in site A, which had been attributed to the wastewater and sewer deposits in combined sewers. However, the TP concentration in storm sewers of site B did not show an increase but a slight decrease. This phenomenon could be explained in two aspects: (1) the mistakenly discharged wastewater and sewer deposits did not contain a significant amount of phosphorus; (2) the high concentration of TP in RD was partly diluted by the DR and RF.



Fig. 2. Distributions of TP under different drainage systems in six rainfalls.

3.4. Distributions and transport of Zn(II), Cu(II), and Pb (II) under different drainage systems

The distributions of Zn(II), Cu(II), and Pb(II) were presented in Fig. 3. Generally the three heavy metals presented a similar variation pattern, although some differences did exist from the specific point of view. In site A, the Zn(II), Cu(II), and Pb(II) concentrations of RF were 4.7, 2.8, and 2.3 times as that in DR, respectively. The particular high ratio in Zn(II) was probably induced by the erosion of zinc materials in the rooftop. Similarly, in site B, the corresponding ratios for Zn(II), Cu(II), and Pb(II) were 3.8, 3.4, and 2.1, respectively. The zinc and copper materials of the



Fig. 3. Distributions of heavy metals under different drainage systems in six rainfalls: (a) zinc ions, (b) copper ions, and (c) lead ions.

rooftop in site B also lead to the extra increase in Zn (II) and Cu(II) concentrations. With regards to RD in site A, its EMC ratios compared to DR were 8.6, 10.3, and 9.3, respectively for Zn(II), Cu(II), and Pb(II), which were roughly in the same levels. Differently, in site B, the corresponding values were 6.3, 12.1, and 5.1 for Zn(II), Cu(II), and Pb(II), respectively. The extraordinary large ratio in Cu(II) should at least partly result from the cooper materials in the standard and cover of street lamps.

Besides, it was found that the gap in Pb(II) concentrations between site A and B were not as large as that in Zn(II) or Cu(II), especially for DR. Considering vehicle emissions was an significant source for Pb(II) [23,24], the close Pb(II) concentrations in site A and B may be caused by the similar vehicle number and flux. Furthermore, the EMC of three-heavy metals in SO of site A were all lower than RD while higher than RF, indicated that the wastewater and sewer deposits did not contribute an amazingly quantity of heavy metals as ammonia nitrogen or TP to the SO.

3.5. Feasibility of rainwater utilization from different surfaces

In order to reveal the feasibility of rainwater utilization in the two studied sites, the obtained water quality of different stormwater runoff was compared with the "Standards for Drinking Water Quality (SDWQ)" in China [25] and presented in Table 2. According to SDWQ, for central water supply, no limit was specially set for NO_2^- and TP, except that several phosphorous pesticides such as malathion, parathion, and methyl parathion were solely restricted. Considering the rainwater was commonly collected and stored in rainwater reservoirs or tanks, the EMC of pollutants would be more reliable than instantaneous concentrations in evaluating the possibility of rainwater reuse for drinking water or household water.

After the EMC of nutrients and heavy metals selected in this research were compared with the SDWQ, the results were showed in Table 3. In site A, due to the severe pollution of the atmosphere, especially in ammonia and nitrate nitrogen, none of the DR or surface runoff could be harvested and reused as drinking water resource directly. From the viewpoint of feasibility and cost versus benefit, the RF might be pre-treated and applied as potential drinking water resource if necessary. But the NH₄⁺-N, and NO₃⁻-N removal efficiencies must reach at least 33.8 and 50.8%, respectively before rainwater reuse. Considering the presence of first flush in most of the sampling processes, abandoning the initial heavily-polluted

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Site	Sources	NH_4^+ -N	NO_3^-	NO_2^-	TP	Zn(II)	Cu(II)	Pb(II)
A	DR	0.56	9.29	0.51	0.16	0.027	0.040	0.002
	RF	0.75	24.15	1.15	0.50	0.127	0.010	0.005
	RD	2.71	10.01	0.72	0.92	0.233	0.037	0.019
	SO	4.27	12.27	0.75	2.73	0.173	0.025	0.009
В	DR	0.25	2.36	0.23	0.04	0.013	0.001	0.002
	RF	0.32	4.38	0.38	0.08	0.048	0.005	0.003
	RD	1.48	2.84	0.31	0.47	0.080	0.016	0.008
	SO	2.09	3.40	0.35	0.36	0.070	0.011	0.006
Standards	[25]	0.5	10	/	/	1	1	0.01

Table 2 Average EMC of different sources in six rainfalls (mg/L)

Note: "/" means no limit was set for this parameter in the "Standards for Drinking Water Quality" in China.

Table 3 Feasibility evaluation of rainwater utilization from different sources of two sites

Site	Category	NH_4^+ -N	NO_3^N	Zn(II)	Cu(II)	Pb(II)	Comprehensive Evaluation
A	DR	×	\checkmark				×
	RF	×	×				×
	RD	×	×			×	×
	SO	×	×			\checkmark	×
В	DR	\checkmark	\checkmark				\checkmark
	RF						
	RD	×					×
	SO	×					×

Note: " $\sqrt{''}$ means below the limit and qualified for potential drinking water resource while " \times " means the opposite.

runoff may decrease the EMC of pollutants to a certain extent [26]. Besides, it was also recommended to clean the roof surface frequently in dry weather, which would undoubtedly weaken the influence of dry deposition to the water quality of RF.

As for site B, DR, and RF could be utilized as potential drinking water supply from the perspective of the studied parameters. This should be mainly owing to the comparatively clean atmosphere in the new district. If pathogens and other contaminants also meet the SDWQ, the DR and RF in site B would be ideal drinking water source. However, RD and SO were not qualified due to the excess ammonia nitrogen mainly from anthropic activities. From the comprehensive evaluation of two sites (Table 2), it could be concluded that the air quality is a significant influence factor and reference index in the decisionmaking of rainwater harvesting and reuse.

Apart from being applied as drinking water, the rainwater also had many household uses, such as washing cars, flushing the toilet, and irrigating the green space [27,28]. Besides, recharging the surface water and ground water were also alternatives for rainwater utilization [29]. Understanding the distributions and transport of typical contaminants in different urban stormwater runoff would provide important assistance and support in rainwater reuse.

4. Conclusions

- The anthropic activities contributed adequately ammonia nitrogen and TP to the RD and SO, especially in site A with combined drainage system.
- (2) Zn(II), Cu(II), and Pb(II) showed a similar variation pattern, although there were some small differences owing to the erosion of metal materials from rooftops and street lamps.
- (3) Air quality was found to be a significant influence factor and reference index in the decisionmaking of rainwater harvesting and reuse.
- (4) The RF in site B could be harvested and utilized for potential drinking water supply from the perspective of the studied parameters. While, at least 33.8% of NH₄⁺-N and 50.8% of NO₃⁻-N in RF must be removed before reuse.

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