**RESEARCH ARTICLE** 



# Comparison between snowmelt-runoff and rainfall-runoff nonpoint source pollution in a typical urban catchment in Beijing, China

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Abstract As a climate-driven event, nonpoint source (NPS) pollution is caused by rainfall- or snowmelt-runoff processes; however, few studies have compared the characteristics and mechanisms of these two kinds of NPS processes. In this study, three factors relating to urban NPS, including surface dust, snowmelt, and rainfall-runoff processes, were analyzed comprehensively by both field sampling and laboratory experiments. The seasonal variation and leaching characteristics of pollutants in surface dust were explored, and the runoff quality of snowmelt NPS and rainfall NPS were compared. The results indicated that dusts are the main sources of urban NPS and more pollutants are deposited in dust samples during winter and spring. However, pollutants in surface dust showed a low leaching ratio, which indicated most NPS pollutants would be carried as particulate forms. Compared to surface layer, underlying snow contained higher chemical oxygen demand, total suspended solids (TSS), Cu, Fe, Mn, and Pb concentrations, while the event mean concentration of most pollutants in snowmelt tended to be higher in roads. Moreover, the TSS and heavy metal content of snowmelt NPS was always higher than those of rainfall NPS, which indicated the importance of controlling snowmelt pollution for effective water quality management.

**Keywords** Nonpoint source pollution · Urban · Snowmelt · Rainfall-runoff · Dust · Leaching experiment

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#### Introduction

Urbanization has accelerated in recent years worldwide, and cropland and woodlands have been converted to roads, buildings, and other paved areas (Jia et al. 2013). This development has increased the impermeability of urban surfaces and changed the build-up and wash-off processes of nonpoint source (NPS) pollutants (Burns et al. 2015). As a climatedriven event, NPS pollution can be caused by rainfall- or snowmelt-runoff processes over and through the ground, which would concentrate natural and anthropogenically deposited pollutants and transport them to receiving water bodies, such as rivers, lakes, seas, or groundwater (Hu and Huang 2014). Studies have indicated that urban NPS pollution is one of the main sources of surface water pollution, and excess loadings of urban NPS pollutants can deteriorate surrounding water bodies (Huang et al. 2013) and cause serious problems, such as eutrophication or toxicity, as well as reducing the quality of instream water habitats (Fučík et al. 2012). Typically, NPS processes involve rainfall-runoff and snowmelt-runoff, while their driving force is wet deposition, which includes rainfall and snowfall inputs. During the wet season, rainfall NPS originates from direct rainfall-runoff processes. During the winter season, snowmelt NPS results from the natural melting of snow into surface water.

Unlike the soil in agricultural areas, surface dust is identified as a key source of urban NPS. Research has shown that the concentration of NPS pollutants increases with an increase in surface dust, indicating that pollutants are concentrated in dust (Li and Zhang 2015). Thus, the seasonal distribution of pollutants in dust has an impact of the seasonal variation of NPS. Li (2013) has concluded that rainfall may contribute to the seasonal change of the level of Pb and As in the dusts. Seasonal influences on road runoff are typical for cold climates (Hilliges et al. 2017). In northern Europe, snow can

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accumulate pollutants in dust during winter, releasing snow storage during spring snowmelt, leading to momentarily very high pollutant loads if snowmelt occurs during a short period in spring. In northern Sweden, eight times higher concentrations and five times higher loads of particles were measured during the snowmelt period compared to warmer periods (Westerlund and Viklander 2006). Other studies have also examined the relationship between stormwater quality and the grain-size distribution of surface dust. Zhao et al. (2010) analyzed the grain-size distribution of road-deposited sediment from Beijing and found that road-deposited sediment with a smaller grain size had a higher metal concentration. Chow et al. (2015) also concluded that fine particles increasingly dominate the pollutant loading on road surfaces. Shen et al. (2016) investigated the influence of grain-size distribution on the stormwater quality, which suggested that the finer grain-size fraction (<38 µm) may be the main source of dissolved pollutants, while coarser particles (74-300 µm) may contribute to solid phases. However, few researchers have analyzed the mechanism of NPS caused by dust from the perspective of wash-off dynamics, which is a possible direction because runoff pollution is closely related to the dissolution process. Thus, the leaching mechanism of pollutants from dust to rainfall-runoff or snowmelt should be further discussed.

In addition, the majority of snow events occur in northern cities in winter. Studies have shown that winter heating, urban traffic, and industrial production are the main source of snow NPS, and a large amount of pollutants can be accumulated in the snow, which would lead to water pollution as the snow melts into the surface water or groundwater (Moghadas et al. 2015; Siudek 2016). Previous studies on snow and snowmelt mainly focused on three aspects: exporting mechanism, model simulation, and control measures. For example, early research mainly focused on changes in snowmelt with location, the pollutant amount in snow, and the effects of a snow melting agent on snowmelt (Reinosdotter and Viklander 2005). Kuoppamäki et al. (2014) analyzed a broad array of contaminants in snowpacks along roads with different traffic intensities and found that pollutants were higher next to highintensity roads compared to low-intensity roads. Siudek et al. (2015) observed a large inter-seasonal variability depending on anthropogenic emissions, depositional processes, and meteorological conditions in the snow cover from an urbanized region in central Poland. Rivett et al. (2016) quantitatively demonstrated that winter leakage to groundwater of a losing stream receiving highway storm-sewer discharges containing deicing salt may constitute a significant linesource of chloride to the underlying aquifer. Models have also been developed to forecast the effect of snowmelt on NPS (Tahir et al. 2011; Valeo and Ho 2004). Additionally, some authors have explored snowmelt NPS management measures (Bartlett et al. 2012; Oberts 1994). Muthanna et al. (2007) examined the treatment of roadside snowmelt on the bioretention with respect to pollutant removal, pollutant pathways, and major sinks. Kratky et al. (2017) concluded that contaminants that were removed via filtration were also not impacted by cold climates, while dissolved contaminants, nutrients, and organics are significantly more variable in their ability to be removed or degraded via bioretention in colder temperatures. However, the study of urban snowmelt NPS was mainly concentrated in cold climate regions. Although those studies discussed the characteristics of snow and snowmelt from different viewpoints, there is still a lack of systematic research into snowmelt NPS from source to output in urban areas.

Furthermore, the characteristics and mechanisms of snowmelt NPS and rainfall NPS are different. Comparatively, rainfall NPS has attracted more attention, and the management of water quality has mainly focused on rainfall NPS. Previous studies have identified the characteristics of rainfall NPS and quantified the impacts of land use (Cheema et al. 2017; He et al. 2010; Rocher et al. 2004); however, the source and runoff quality of snowmelt NPS and rainfall NPS are different, and the methods used to manage rainfall NPS are not applicable to snowmelt NPS. Thus, it is also necessary to explore the runoff quality of snowmelt NPS and rainfall NPS. At present, only a few researchers have analyzed the characteristics of rainfall NPS and snowmelt NPS on a regional scale. Westerlund and Viklander (2006) found much higher loadings of particles and associated metals during the snowmelt period compared to the rain period in Sweden. Li et al. (2012) analyzed and compared snowmelt NPS with rainfall NPS in Xi'an city, China, and discovered that snowmelt NPS had an initial flushing effect to a certain degree and that TP and Zn concentrations in the snowmelt were higher than those in the rainfall-runoff. Urban runoff characteristics are very site specific and local environmental and social-economic conditions vary from location to location. Therefore, local, or at least regional, guides would be most helpful. For example, China has been implementing the Sponge City Construction since 2014, which mainly focuses on urban drainage system and NPS control (Ren et al. 2017). However, localized technical guidance is still not available for many designated sponge cities because the mechanism of urban NPS is not systematically researched, which limited the development of control measures (Jia et al. 2017). Furthermore, comparison of the runoff quality of snowmelt NPS and rainfall NPS are limited yet.

One of the objectives of the present study is to determine the characteristics of snowmelt NPS and rainfall NPS. Specifically, our objectives are (1) to study the seasonal variation and leaching characteristics of pollutants in surface dust, (2) to analyze the pollution of snow and snowmelt NPS, and (3) to compare the runoff quality of snowmelt NPS and rainfall NPS. A typical community in Beijing, China, was selected for this study. The current work contributes insights into the characteristics of NPS pollution in regions of medium latitudes with monsoon climates.

# Materials and methods

#### Site description

In this study, we chose Beijing Normal University (BNU) as the study area to explore the characteristics of snowmelt NPS. Considering the city's dense population, frequent traffic activity, and typical impervious surfaces, we chose the study area around BNU (Fig. 1), which is located between the 2nd Ring Road and the 3rd Ring Road and belongs to the central area. Previously, our group conducted a series of studies on rainfall NPS in this area. BNU has a typical semi-humid continental monsoon climate, with a hot and rainy summer, cold and dry winter, and short spring and autumn.

To examine the NPS caused by rainfall and snowmelt, it is necessary to select representative typical functional areas or land types as sampling sites by analyzing their functional zoning and land use. Through comprehensive consideration of the impact of land use and geographical location on surface water pollution, our research intends to monitor the surface rainfall-runoff and snowmelt at three sampling sites, including roads from an office area and a trafficheavy area and the roof of a building. In addition, the pollutant concentrations in surface dust were closely related to the concentration of pollutants in the runoff. Previous studies have found that surface dust on impervious surfaces (roads and roofs) was more easily washed off by runoff (Barbosa et al. 2012; Chua et al. 2009; Egodawatta et al. 2007; Jia et al. 2015), and thus, surface dust was also collected to analyze runoff pollution. The details and location of each sampling point is shown in Table 1.

#### Surface dust sample collection and analysis

#### Dust collection

The study was conducted from June 2012 to February 2014. The sampling of rainfall-runoff was mainly performed during the rainy summer season, while snow sampling was carried out at the end of 2012 and early 2013. In addition, surface dust was collected monthly during 2013. The samples are described in detail in Table 2. A total of 548 samples were collected and analyzed in this study, and more than seven dry days preceded sampling to ensure that the build-up process had stabilized (Wicke et al. 2012). The samples were collected over an area of 1 m  $\times$  1 m at each sampling site. The samples were obtained by manual cleaning with a brush and dustpan. The

entire process from sample collection to processing ensured no exposure to metal tools. In addition, three parallel samples were collected at each sampling point.

# Leaching analysis of pollutants in dust

In this study, an immersion experiment was designed to study the effect of surface particles on the dissolution of pollutants (heavy metals and nutrients) at certain intervals. Considering the time span from runoff generation to output, the duration of the immersion experiment was 5 h. According to our previous study (Ouyang et al. 2015), the pH of rainwater in Beijing is basically neutral, and thus, neutral water was used in this study. The dust samples (5 g) were transferred to clean 100-mL conical flasks and numbered. Then, 50 mL of purified water was added to the conical flasks to give a solid-to-liquid ratio of 1:10. Water samples were treated as follows: shaking at 180 rpm for a certain time interval (0.5, 1, 2, 3, 4, 5 h), centrifugation at 4000 rpm for 15 min, and finally extraction of the supernatant to a polyethylene bottle until use. Additionally, blank and parallel experiments were conducted to minimize disturbances and improve accuracy. In this study, heavy metals (Cu, Fe, Mn, Zn, and Pb) and nutrients (total phosphorus and total nitrogen) were analyzed. Specifically, the heavy metals and nutrients were measured by spectrophotometric methods, which referred the standard methods for water and wastewater monitoring and analysis (fourth edition) (SEPA 2002). To evaluate pollutant leaching, the leaching ratio was introduced and calculated as follows (Xu et al. 2017):

$$E = \frac{Li(t)}{Mi} \times 100\% \tag{1}$$

where *E* is leaching ratio of pollutants in water,  $L_i$  is the amount of the *i*th element extracted from dust in water in *t* (mg/kg),  $M_i$  is the amount of the *i*th element contained in the raw dust mixture (mg/kg), and *t* is the certain time.

#### Snow and rainfall-runoff sample collection and analysis

#### Sample collection

Snows were sampled at the end of 2012 and early 2013, and all samples were collected within 24 h after snowfall. The details are shown in Table 2. Three samples were obtained from each site, including the surface snow, underlying snow, and total snow. Specifically, the surface snow samples were taken 1 cm from the surface of the snow, while underlying snow samples were taken 1 cm from the bottom of the snow. The total snow samples were taken from the surface to the bottom at each site.

Fig. 1 The study area and locations of sampling sites; roads and roofs are also shown; circle represents the sampling sites of rain and snow; triangle represents the sampling sites of dust



The samples were taken during snowfall and then sealed in a polyethylene bag. The collected samples melted naturally in the laboratory and were then poured into polythene bottles and stored at 4 °C until subsequent analysis.

Rainfall-runoff samples were mainly collected in the rainy season. These samples were collected at set time intervals after runoff occurred. More details of rainfall-runoff samples can be found in the work of Shen et al. (2016).

### Sample analysis

In this study, pollutants in the snowmelt and rainfall-runoff were analyzed, including the pH, chemical oxygen demand (COD), total suspended solids (TSS), total nitrogen (TN), total phosphorus (TP), and metal elements (Cu, Fe, Mn, Zn, and Pb). All samples were analyzed following the standard methods for water and wastewater monitoring and analysis

**Table 1**Brief information ofsampling sites, land typematerials, and remarks in BNU

Sampling	sites	Materials	Remarks			
Roof	Gym	Asphalt and shingle	Flat roof, with total area of 30 m <sup>2</sup>			
	Laboratory building	Asphalt and shingle	Flat roof, with total area of 565.7 $m^2$			
	Office building	Asphalt and shingle	Flat roof, with total area of 785.8 m <sup>2</sup>			
Road	North 3rd Ring Road	Asphalt	Main road with high density traffic volume (110,000 vehicles/day)			
	Xingtan Road	Asphalt	Side road with medium density traffic volume (17,000 vehicles/day)			

**Table 2**Description of the sampling periods for dust, rainfall, andsnow in BNU

Sample ti	me	Dust	Rainfall	Snow
2012	November	$4  imes 3^{a}$	6 × 11	
	December	$4 \times 3$		$4 \times 8$
2013	January	$4 \times 3$		$7 \times 8$
	February	$4 \times 3$		$4 \times 9$
	March	$4 \times 3$	$3 \times 10$	
	April	$4 \times 3$		
	May	$4 \times 3$		
	June		5 × 12	
	July		6 × 12	
	August	$4 \times 3$		
	September	$4 \times 3$	5 × 13	
2014	February	$4 \times 3$	$4 \times 12$	
Total		120	341	87

 $^{a}(4 \times 3)$  indicates the number of sampling points multiplied by the number of samples collected at each sample point in the table. Specifically, the number of sampling sites was 4 for dust and 5 for rainfall and snow. However, the number of sampling sites for each event was uncertain due to the limitations of the human resources

(fourth edition) (SEPA 2002). More concretely, the COD was measured by using potassium chromate volumetric analysis method; the heavy metals and nutrients were measured by spectrophotometric methods.

The event mean concentration (EMC) was used to compare the pollutant concentrations in snowmelt NPS and rainfall NPS. The EMC is a flow-weighted average concentration computed according to the following equation (Peng et al. 2016):

$$EMC = \frac{M}{V} = \frac{\int_0^T C_t Q_t dt}{\int_0^T Q_t dt}$$
(2)

where EMC is the event mean concentration (mg/L), M is the total amount of pollutant discharged from a snow or rainfall event, V is the sample volume of rainfall-runoff and natural snowmelt,  $C_t$  is the concentration of pollutants at time t (mg/L),  $Q_t$  is the flow rate of runoff at time t (m<sup>3</sup>/s), and t is the time period.

Furthermore, the EMCs were statistically processed using the SPSS software to analyze the changes in pollutants in the snowmelt and rainfall-runoff from roofs and roads.

# **Results and discussion**

# Seasonal variation and leaching of pollutants in surface dust

The seasonal distributions of heavy metals and nutrients in the surface dust from different sampling sites are shown in Fig. 2.

Overall, the concentrations of nitrogen, phosphorus, and heavy metals were higher in the winter and spring than in the summer and autumn, which is generally consistent with previous studies (Cao et al. 2015; Li and Zhang 2015). Because of the sub-humid, warm, continental monsoon climate, the windy weather in spring leads to an increased amount of dust particles on surfaces. In addition, Beijing has experienced rapid urbanization. Population activities, transportation, and winter heating all contribute to the collection of more surface dust. In addition, the accumulation of surface dust increased during the longer dry periods due to the reduced precipitation in winter. Xiong et al. (2015) studied the distribution characteristics of trace element and heavy metal pollution from November 2013 to March 2014 and indicated that sources of winter dust fall in Beijing city mainly came from the Earth's crust (including road dust, construction dust, and remotely transmitted dust) and the burning of fossil fuels (vehicle emissions, coal combustion, biomass combustion, and industrial processes). Furthermore, dust was collected from roads in a trafficheavy area and an office area and from a roof in a residential area. According to Fig. 2, the pollutants in dust from different underlying surfaces have significant seasonal differences, which indicates that the temporal deposition of pollutants in surface dust are different, which might be the primary cause of the difference between rainfall NPS and snowmelt NPS. In general, the temporal changes in the concentrations of TN and Cu are obvious, while the concentration of Fe in surface dust does not show a significant change in different seasons. In addition, TN had highest value in winter, which indicates that TN pollution requires more attention in winter. By contrast, TP and Cu had higher concentration in spring, which suggests that TP and Cu pollution require more attention in spring.

To explore the impact of dust on NPS, the leaching concentration of dust was further analyzed, and for this, the dust sample from the North 3rd Ring Road collected on March 19, 2013, was used. The variation in the concentration of dissolved pollutants in surface dust over time is provided in Fig. 3, and the leaching ratio of pollutants in the dust, such as heavy metals and nutrients (TN and TP), was analyzed at different time periods. According to Fig. 3, the heavy metal concentration experienced significant fluctuations at set intervals, which shows that the reaction time has an obvious effect on the leaching concentration of heavy metals. Overall, the leaching concentration of the majority of the metals decreased after initially increasing and finally tended to stabilize, except for Cu and Zn, which showed an overall downward trend. The leaching ratios of the various heavy metals varied greatly. Cu and Zn were easily desorbed compared with the other metals (Mishra 2014). The desorption of soluble Cu and Zn to colloidal or insoluble particles or the complexation of soluble Cu and Zn with other pollutants in the liquid phase may decrease





Fig. 2 Seasonal distribution of Cu, Fe, TN, and TP in surface dust

the solubility of heavy metals. Other heavy metals (Ni, Fe, Cr, and Pb), which were weakly adsorbed to the particles, were rapidly desorbed and released. Subsequently, some heavy metal ions were released by ion exchange and complexation with soluble organic matter. With a change in the soaking environment, the combination of desorption, ion exchange, and complexation stabilized the concentration of heavy metals. Liu et al. (2014) investigated the effects of reaction time in the leaching process on the leaching ratio for the Cr–Fe alloy and suggested that Cr present on the surface of the Cr–Fe alloy powder was easily passivated as  $Cr_2O_3$ , which caused the surface reaction rate to decrease and stabilize after the optimized reaction time. However, the concentrations of TN and TP in the liquid phase showed significant changes over time, and their trends were similar over time. The concentrations of TN and TP in the liquid phase first increased, then decreased and then reached higher values after 1 and 3 h, respectively. Therefore, TN and TP require more time to



Fig. 3 Variation in the leaching concentration of pollutants in surface dust with leaching time

 Table 3
 Leaching concentration

 and ratio of heavy metals in
 surface dust at different times

	Extractio	Extraction from dust										
	0.5 h	0.5 h		1 h		2 h		3 h		4 h		
	mg/L	%	mg/L	%	mg/L	%	mg/L	%	mg/L	%		
Cu	0.35	1.85	0.35	1.87	0.31	1.62	0.30	1.58	0.28	1.49		
Mn	0.08	0.20	0.07	0.18	0.07	0.18	0.08	0.19	0.08	0.20		
Pb	0.010	0.12	0.011	0.14	0.010	0.12	0.008	0.10	0.008	0.10		
Zn	0.16	0.61	0.12	0.48	0.10	0.39	0.10	0.39	0.08	0.32		
Fe	0.15	0.01	0.17	0.01	0.16	0.01	0.18	0.01	0.17	0.01		
ТР	0.66	0.69	1.55	1.62	1.21	1.27	1.66	1.73	1.46	1.52		
TN	30.8	18.4	34.8	20.7	33.8	20.1	44.1	26.3	37.0	22.1		

stabilize during leaching (Desboeufs et al. 2014). The above results indicate that the exporting processes of NPS are greatly influenced by the types of dust and pollutants (Liu et al. 2015).

Table 3 lists the detailed leaching concentrations of the pollutants. As shown in Table 3, the leaching ratio of the various heavy metals varied greatly. The ratio of heavy metals exhibited the following order: Cu > Zn > Mn > Pb > Fe. Specifically, compared to other heavy metals, Cu had the highest leaching ratio, while Fe had the lowest leaching ratio, reaching 1.87% and 0.01%, respectively. However, TN and TP had higher leaching ratios than the majority of the heavy metals. In addition, the leaching ratio of TN was much greater than that of TP, and both reached their highest values within 3 h of less than 26.3% and 1.73%, respectively. Therefore, the leaching ratio of pollutants was very low, which is generally consistent with previous research. Dissolution studies on street dust have suggested that the elements are slightly soluble under normal conditions (around neutral pH) (Joshi et al. 2009). The results indicated that the amount of soluble pollutants in dust was low, and most pollutants entered the runoff in particulate form. This finding provides background for the development of monitoring methods and runoff pollution treatment.

## Characteristics of pollutants in snow and snowmelt

Table 4 shows the characteristics of pollutants in underlying snow and surface snow. In this study, three snow events were monitored to analyze the pollutant distribution. As can be clearly seen from Table 4, a great difference was observed for pollutants in the surface snow and underlying snow for the same snow event. Apparently, the concentrations of TSS, COD, Cu, Fe, Mn, and Pb were higher in the underlying snow than in the surface snow. For example, the COD in the underlying snow was approximately four times that in the surface snow. However, the concentrations of NH<sub>3</sub>-N, NO<sub>3</sub>-N, and Zn in the surface snow were higher than those in the underlying snow. These results indicated that the sources of these pollutants were different. The reasons for this situation are as follows. On one hand, the underlying snow adsorbed more surface dust compared with the surface snow, and on the other hand, the concentrations were related to the atmospheric

Table 4 Pollutant concentrations in surface snow and underlying snow for different snow events (all values in mg/L except for pH)

	2012/12/14		2013/1/21		2013/2/5		Mean		
	Surface	Underlying	Surface	Underlying	Surface	Underlying	Surface	Underlying	
pН	8.71	7.79	9.12	9.01	6.57	6.28	8.13	7.69	
COD	48.0	75.0	20.0	130.0	135.0	470.0	67.7	225.0	
TSS	16.0	66.5	82.0	64.0	17.0	66.0	38.3	65.5	
NH <sub>4</sub> -N	0.61	4.36	24.03	20.86	12.39	10.05	12.34	11.76	
NO <sub>3</sub> -N	0.24	0.41	2.72	1.48	1.50	1.45	1.49	1.11	
ТР	0.011	0.014	0.031	0.023	0.019	0.022	0.021	0.020	
Cu	0.002	0.010	0.006	0.005	0.015	0.013	0.008	0.009	
Fe	0.018	0.079	0.006	0.013	0.028	0.016	0.017	0.036	
Mn	0.007	0.032	0.059	0.040	0.029	0.036	0.031	0.036	
Zn	0.088	0.206	0.163	0.139	0.539	0.376	0.263	0.240	
Pb	0.0007	0.0106	0.0018	0.0012	0.0030	0.0036	0.0018	0.0051	

Table 5 Pollutant concentrations of snowmelt at roofs (GF, LF, and OF) and roads (TD and XD) for different snow events (mg/L)

	2012/12/14			2013/01/21					2013/02/05			Mean
	GF	LF	OF	GF	LF	OF	TD	XD	LF	TD	XD	
pН	7.55	8.43	8.36	7.74	8.33	7.61	8.31	8.59	7.46	7.19	7.50	7.92
COD	130.6	143.8	130	345.7	470	363.3	960	570	778.3	1212.5	1142.2	567.85
TSS	41.8	86.5	50.4	168.9	142.3	122	1268	823.5	140.3	1236.7	215.8	390.56
NH <sub>4</sub> -N	12.78	9.99	11.88	34.11	36.55	41.88	66.13	52.4	32.93	28.62	22.83	31.83
NO <sub>3</sub> -N	5.3	13.57	2.35	9.26	14.44	14.56	15.51	10.88	14.87	8.26	6.47	10.5
TN	-	-	-	43.56	51.96	56.64	85.73	63.28	48.98	38.17	29.51	52.23
ТР	0.019	0.033	0.018	0.043	0.083	0.057	0.765	0.726	0.085	0.23	0.17	0.203
$SO4^{2-}$	-	-	-	153	227.5	185	635.3	357.5	268.1	187	168.4	272.73
$Cl^-$	_	_	_	140	141	157	59,851	23,986	120	7369	17,743	13,688
Cu	0.02	0.029	0.016	0.045	0.041	0.084	0.225	0.143	0.043	0.173	0.068	0.081
Fe	0.173	0.027	0.042	0.084	0.071	0.222	0.117	0.102	0.042	0.044	0.024	0.086
Mn	0.139	0.08	0.074	0.425	0.2	0.531	0.439	0.529	0.079	0.287	0.324	0.282
Zn	0.353	11.856	0.745	6.883	14.095	3.303	0.244	0.253	11.01	0.283	0.208	4.476
Pb	0.0047	0.0054	0.0034	0.0069	0.0165	0.0188	0.027	0.0061	0.0127	0.0115	0.0049	0.0107

"-" indicates the species was not detected

environment. Surface snow more easily absorbs NH<sub>3</sub>-N from air (Xia et al. 2014). These results indicated that snow provides a potential source for NPS.

It is generally known that the accumulation and transformation of pollutants in a snow cover have negative effects on groundwater, terrestrial environments, and human health (Dossi et al. 2007; Siudek et al. 2015). To investigate the impact of snow on urban NPS, we also conducted a series of snowmelt monitoring tests during the snow events. The amount of sampling points and samples collected during each snowfall event were different due to the difficulty of snow collection. The detailed snowmelt monitoring results are given in Table 5. Specifically, GF, LF, and OF represented the roof of a gym, a laboratory, and an office building, while TD and XD represented the 3rd Ring Road and Xingtan Road. As can be clearly seen from Table 5, the concentration of pollutants in the snowmelt from the various snow events varied greatly. Through statistical analysis, we discovered that the COD on the roof was 130-143.8 mg/L on December 14, 2012, but was 345.7-470 mg/L on January 21, 2013. The TSS concentration on the roof ranged from 41.8 to 86.5 mg/L on December 14, 2012, but ranged from 122 to 168.9 mg/L on January 21, 2013. The content of NH<sub>4</sub>-N, NO<sub>3</sub>-N, and TP on the roof was high in the snowmelt on January 21, 2013, compared with that on December 14, 2012. The concentration of pollutants in the road also varied greatly with the various snow events. For example, the COD ranged from 570 to 960 mg/L on January 21, 2013, but ranged from 1142.2 to 1212.5 mg/L on February 5, 2013.

Furthermore, the concentration of pollutants in the same snow event is remarkably different at different sampling points. Specifically, the mean values of NH<sub>4</sub>-N and NO<sub>3</sub>-N from the roof were similar to that from the road for snowmelt, while COD, TSS, TP,  $SO_4^{2-}$ , and Cl<sup>-</sup> in the road were higher than those on the roof. In particular, the concentration of Cl<sup>-</sup> in the road was two orders of magnitude higher than that on the roof due to the administration of road salt in winter (Zhu et al. 2012). In addition, the traffic density also has an impact on snow NPS. In Table 1, TD is the main road with high traffic density (110,000 vehicles/day), while XD is a side road with a medium traffic density (17,000 vehicles/day). As seen from Table 5, all pollutants from TD were higher than that from XD, except for Cl<sup>-</sup>. The results indicate that snowmelt from the main road contained more pollutants than that from the side road (Sun et al. 2016). However, Cl<sup>-</sup> mainly came from snow melting agents, and its concentration was greatly affected by the amount of snow melting agent administered. In Beijing, the snow remover was mainly based on the machine snow plow, and partially road salt from 2012, which caused the uncertainty of  $Cl^{-}$  in road.

Based on the analysis in Table 5, the concentration of metals in the snowmelt on the roof was high on January 21, 2013, compared with the snowmelt on December 14, 2012, except for Fe in GF. Considering the similarity of GF, LF, and OF, the outliers may be caused by errors in the sampling and monitoring processes. Additionally, the concentrations of metals in the snowmelt from the roads



Fig. 4 Comparison of the pollutants from snowmelt NPS and rainfall NPS; blue and green represent the pollutant from roads and roofs, respectively

were higher than those from the roofs, except for Zn. Furthermore, most pollutants from TD (main road) were higher than that from XD (side road). In addition, the average concentrations of Cu, Zn, and Pb in snow from Beijing were much higher than those from Cerro Colorado in Chile, Poznań in Poland, and Novy Sad in Serbia, located in developed areas (Siudek et al. 2015).

#### Comparison of snowmelt NPS and rainfall NPS

Figure 4 shows the runoff quality of pollutants analyzed by comparing the snowmelt with the rainfall-runoff monitored in the wet season. A box plot was used to intuitively analyze and compare the average and variance of multiple data sets. In this study, we analyzed and compared the difference in the

maximum, minimum, and median concentration of snowmelt NPS and rainfall NPS. The data in the statistical analysis mainly came from different sampling events and varied greatly. The pH in snowmelt NPS from the roads ranged from 7.19 to 8.59, while the pH in rainfall NPS from the roads was 7.72-8.51; the COD of snowmelt NPS and rainfall NPS from the roads ranged from 570.0 to 1212.5 mg/L and 219.47 to 760.8 mg/L, respectively; the TSS of snowmelt NPS and rainfall NPS from the roads was 215.0-1236.67 mg/L and 295.5-608.74 mg/L, respectively; the NH<sub>3</sub>-N, NO<sub>3</sub>-N, TP, Cu, Fe, and Mn in snowmelt NPS from the roads was 22.83-66.13 mg/L, 6.47-15.51 mg/L, 0.17-0.765 mg/L, 0.068-0.225 mg/L, 0.024-0.117 mg/L, and 0.287-0.529 mg/L, respectively; the NH<sub>3</sub>-N, NO<sub>3</sub>-N, TP, Cu, Fe, and Mn in rainfall NPS from the roads was 11.00-82.44 mg/L, 2.52-25.29 mg/ L, 0.028-2.859 mg/L, 0.019-0.285 mg/L, 0.014-0.46 mg/L, and 0.065–0.288 mg/L, respectively. The COD, TSS, NH<sub>3</sub>-N, NO<sub>3</sub>-N, TP, Cu, Fe, and Mn in snowmelt NPS from the roofs was 130.0-778.33 mg/L, 50.4-142.25 mg/L, 9.99-41.88 mg/ L, 2.35-14.87 mg/L, 0.018-0.085 mg/L, 0.016-0.084 mg/L, 0.027-0.222 mg/L, and 0.074-0.531 mg/L, respectively; on the other hand, the COD, TSS, NH<sub>3</sub>-N, NO<sub>3</sub>-N, TP, Cu, Fe, and Mn in rainfall NPS from the roofs was 64.6-341.4 mg/L, 16.36-87.5 mg/L, 11.96-55.96 mg/L, 4.53-46.03 mg/L, 0.017-0.163 mg/L, 0.007-0.056 mg/L, 0.039-0.335 mg/L, and 0.042-0.211 mg/L, respectively. Thus, the variation of NH<sub>3</sub>-N, NO<sub>3</sub>-N, TP, Cu, and Fe in rainfall NPS was larger than that in snowmelt NPS, which was related to the varying number of preceding dry days. In the summer, rainfall events were frequent and random, and the pollutants that were built up in impervious areas varied with different numbers of preceding dry days. In addition, the maximum of each pollutant in rainfall NPS was larger than that in snowmelt, except for COD, TSS, and NH<sub>3</sub>-N, which had higher values in snowmelt NPS.

In this study, we also analyzed the median EMC. The results indicated that there was a difference between snowmelt NPS and rainfall NPS. According to Fig. 4, the mean value of the pH was higher in snowmelt NPS compared with rainfall NPS. The main reason for this difference is that snow melting agents in China often contained potassium acetate with alkalescence, which led to a higher pH in the snowmelt. Additionally, the snow is exposed to particulates for a longer time when they fall and melt, which is one of the main reasons for the high TSS concentration. Sun et al. (2016) demonstrated that TSS and COD were greatly affected by human activities during the period of snowmelt. In this study, TP was more serious in rainfall NPS than in snowmelt NPS. Moreover, some studies have found that the EMCs of heavy metals in snowmelt NPS are higher than those in rainfall NPS, which is in agreement with our results (Hilliges et al. 2017; Valtanen et al. 2015). As shown in Fig. 2, the accumulation of nitrogen, phosphorus, and heavy metals in dust were higher in winter and spring than in summer and autumn. In addition, these results are greatly related to the environmental conditions at the time. Snowfall carried more pollutants than rainfall, partly because of the long contact time during snow events but also because more pollutants are carried by melting snow, which has a large potential for spreading pollution. Therefore, pollution from snowmelt cannot be ignored.

# Conclusion

In this study, three factors relating to urban NPS, including surface dust, snowmelt, and rainfall-runoff processes, were comprehensively analyzed, and the snowmelt NPS and rainfall NPS were compared. Based on the results, dust deposition contributed more pollutants in winter and spring, and these built-up pollutants showed low leaching ratios, indicating that most NPS pollutants were particulate forms. Underlying snow has higher COD, TSS, Cu, Fe, Mn, and Pb concentrations than surface snow, while higher EMCs of most pollutants were observed in road snowmelt. In addition, the snowmelt NPS had higher COD, TSS, and metal content than rainfall NPS, indicating the importance of controlling snowmelt pollution for effective water quality management.

However, there are also some limitations of this study. Due to the limited experimental conditions, snowmelt NPS was only conducted in the laboratory rather than with snowmelt-runoff in field conditions. In addition, the duration of the immersion experiment was designed as 5 h, considering the time span from runoff generation. It is suggested that this experiment should be longer to obtain stabilized leaching results for N and P.

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#### Compliance with ethical standards

Conflict of interest The authors declare no conflicts of interest.

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