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Influence of surface properties and antecedent environmental conditions on particulate-associated metals in surface runoff

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A B S T R A C T

Particulate-associated trace metals have been regarded as an important pollution source for urban surface runoff. Cd, Pb, Cu, Zn and total solids (TS) washed off two different surfaces (low-elevated facade and road surfaces) under two kinds of antecedent environmental conditions (dry and snow-melting) were determined in this study. Wet-vacuuming sweeping (WVS) and surface washing (SW) methods, representing the maximum pollution potential and common rainfall-induced wash-off condition respectively, were used to collect the particulate matters. The result shows that the wash-off concentrations of trace metals were found in the order of Cd (2.28 ± 2.08 μg/l) < Pb (435.85 ± 412.61 μg/l) < Cu (0.93 ± 0.61 mg/l) < Zn (2.52 ± 2.30 mg/l). The snow-melting process had a considerable influence on the wash-off concentrations of the trace metals on both road and facade surfaces. It reduced >38% and >79% of metals and TS concentrations in the facade surface and road surface runoff respectively. The wash-off concentrations of Cd, Cu, and Zn on the road surface 45–780% higher than those on the facade surfaces. The sensitivity analysis based on the Bayesian network indicates that the wash-off concentrations of metals were mainly dependent on the antecedent environmental conditions or the surface properties while the sampling methods had a minor influence. Therefore, to accurately model the pollutant migration in the surface runoff requires an improving method considering different surfaces and antecedent environment conditions.

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1. Introduction

Urban stormwater runoff affects water quality, water quantity, habitat and biological resources, public health, and the aesthetic appearance of urban waterways [1,2]. It has gained increasing attention and the deteriorated stormwater runoff has been regarded as a leading source of pollutants causing water quality impairment related to human activities in the natural hydrological system [2–4]. The rainfall-induced surface runoff would wash the anthropogenic pollutants of trace metals [5,6], polycyclic aromatic hydrocarbons (PAHs) [7,8], and other toxics off the impervious surface and eventually discharge into the nearby water bodies through storm sewers or combined sewer overflows (CSOs) [9,10]. Among the pollutants found in surface runoff, trace metals are of great concern due to their toxicity, persistence, and abiotic degradation in the environment and bioaccumulation in the food webs [11,12].

In this context, the wash-off process of trace metals during the rainfall event would affect the potential deterioration level of runoff water quality [13,14]. In a conceptual rainfall-runoff model,
the stormwater pollution in the wash-off process is dependent on the build-up process, runoff rate and other influencing variables [1,15]. Antecedent dry weather period (ADP) is commonly recognized as one of the most important influencing variables controlling the pollutants’ build-up process [6,16,17]. Therefore, its impact on the wash-off process was discussed in many studies [18–20]. However, less attention has been addressed to the effect of other antecedent environmental conditions, such as snowpack melting between the snow and precipitation events, on the pollutants’ build-up process and thereafter the occurrence in the surface runoff. The antecedent snowmelt period (ASP) would play an important role at the high latitude zones in early spring when the snow-melting and rainfall-induced runoff occurs and forms a complicated wash-off process. In addition to the antecedent environmental conditions, most of the studies focused on the pollutants’ wash-off on the road and roof surfaces [21–23], but the wash-off process from the building facade was neglected. Therefore, an in-depth study on the wash-off pollutants from the facade could provide a complete understanding of the wash-off process in an urban area and reduce the uncertainty of wash-off results in hydrological models.

Accordingly, in order to assist the best management practice of stormwater pollution mitigation, the focuses of this study are to (i) characterize the particulate-associated trace metals (Cd, Pb, Cu, and Zn), (ii) determine the influence of antecedent environmental conditions (ADP and ASP) and surface properties (road surfaces and low-elevated facade) on the metal pollution levels, and (iii) rank the influential parameters through the sensitivity analysis based on Bayesian network.

2. Materials and methods

2.1. Study area

The road deposited samples were obtained from George-Baehr-Str. (51°01′47″ N, 13°43′24″ E). The sampling street is near Berg str. and Nuenberger str. with a high traffic load which is 13600–15000 vehicles per day [24]. It was selected due to the elevated levels of road deposited sediment adsorbed pollutants (Zhang et al., 2015). All the building facades along the given road are made of the same materials of bricks and mortar, and the road is also paved with a uniform material, asphalt.

2.2. Sample collection

The samples collection was conducted on 25th February and 4th March 2016 with two-day accumulation after the last rainfall or snowfall events for each sampling campaign. More explicitly, there were events with rainfall of 11.9 mm and 1.2 mm on 22nd and 23rd February respectively, while 50 mm snow/0.9 mm rainfall and 40 mm snow/0.7 mm rainfall fell on 1st and 2nd March [25]. On 4th March, snowpack remained on the non-paved pervious areas was collected from the area of 2 m2 adjacent to the road surfaces. In each sampling site, the wash-off samples from the low-elevated facade surface were collected from the area of 2 m height above ground level and 1 m width. The runoff samples from the road surface were obtained from 2 m2 (2 m × 1 m) roadside curb areas, where most sediment is assembled [26,27]. Since there is no standard sampling method for collecting the surface sediment, the samples from both surfaces were collected by two sampling techniques, which are wet vacuuming sweeping (WVS) method [17,28] and surface washing (SW) approach [22,29]. At each sampling site, two sampling methods were performed on the neighboring 2 m2 predefined facade or road surfaces. It was initially uniformly sprayed with 1 L of synthetic rainwater in order to replicate the real wash-off condition.

The synthetic rainwater was made of 1.35 µg/l of NaCl, 1.13 µg/l of HNO3 as well as 1.76 µg/l of H2SO4 with CH3COO- and CH3COONa as buffers [27,30]. NaOH solution was added to adjust the pH of 4.7–5 according to the historical values of the rainwater in Dresden. According to the WVS method, the particles within the analyzed area were collected by a professional vacuum sweeper (Puzzi 100 Super, Kärcher) twice in the different directions, which intend to provide the maximum pollution potential in the surface runoff. However, the SW method collected the runoff from the road at the outlet point, while the facade samples were collected in a plastic basin at the bottom of the surface. It intends to provide the pollution potential in the surface runoff after a normal level of rainfall. Therefore, WVS and SW methods were used to represent the range of potential pollution levels between the maximum to normal in surface runoff.

2.3. Chemical analysis

A total of 48 samples were tested for Cadmium (Cd), Lead (Pb), Copper (Cu), and Zinc (Zn). The collected solutions were initially agitated, acidified with 65% HNO3 and stored at 4 °C in a refrigerator. The 40 ml samples were then filtered through a Sartorius 0.45 µm membrane filter by using a vacuum. The filters and sludge from the previous step were left in an oven at 105 °C for a minimum of 2 h. They were subsequently weighted for total solid (TS) analysis, while the filtrates were analyzed for the total trace metal concentrations.

The concentrations of Cd and Zn were determined using flame furnace atomic absorption spectrometry (FF-AAS). Graphite furnace atomic absorption spectroscopy (GF-AAS) was used to analyze Cd and Pb, which had relatively lower concentrations. The analytical methods and procedures of Cd, Pb, Cu, and Zn were following the German standard Norm DIN EN ISO 5961, DIN38406-6, DIN 38406-7, and DIN 38406-8 respectively. The limits of detection were 0.05 µg/l for Cd, 1.00 µg/l for Pb, 0.01 mg/l for Cu, and 0.01 mg/l for Zn. The limits of quantitation were 0.10 µg/l for Cd, 2.00 µg/l for Pb, 0.02 mg/l for Cu, and 0.02 mg/l for Zn.

2.4. Bayesian network

Bayesian network is a probabilistic graphical model, which visually interprets a causal relationship between variables [31,32]. Comparing with other methods for ranking parameters (e.g. hierarchy cluster analysis), Bayesian network has a stronger capacity on dealing with both discrete and continuous data [33]. It consists of a directed acyclic graph (DAG) and a conditional probability table (CPT), corresponding to qualitative description and quantitative description respectively [34,35]. DAG, composed of the node variable set X = {X1, X2, ..., Xn} and directed edges set, describes the causal relationship between each child node and its parent nodes; CPT quantified the all possible conditional probabilities among these nodes [36]. Bayesian network factorizes the joint probability distribution to several local probability distributions among the nodes:

$$P(X_1, X_2, ..., X_n) = \prod_{i=1}^{n} P(X_i|\pi(X_i))$$

(1)

where P is the probability distribution, \pi(X_i) is the parent node sets of X_i.

The sensitivity of parameters was analyzed through the entropy reduction due to the discrete state of those variables in the simple network. It is a robust local sensitivity analysis used in many
researches [37–39]. The entropy reduction is defined as the expected reduction in uncertainty of the expected real value of the target variable Q due to a finding at an input variable F, and it was calculated as [40,41]:

\[ I = H(Q) - H(Q|F) = \sum_{q} \sum_{f} p(q,f) \log_2 \frac{p(q,f)}{p(q)p(f)} \]

(2)

where \(H(Q)\) is the entropy of Q before any new findings and \(H(Q|F)\) is the entropy of Q after new findings from F. The larger entropy reduction value represents the higher dependency of the parent variable and the target child variable.

The Bayesian network construction and parameter sensitivity analysis were carried out by Netica. The network structure was shown in Fig. 1. Three influential parameters, namely, surface properties, antecedent environmental conditions, and sampling methods were identified to exert influence on the trace metal concentrations. The continuous variables, such as concentrations, were converted into the discrete quantities before the probabilistic inference.

3. Results and DISCUSSIONS

3.1. Wash-off contents of metals and TS

The concentrations of four trace metals (Cd, Pb, Cu, and Zn) collected by the two given methods are displayed in Fig. 2. The results were categorized into different antecedent environmental conditions of 2-day ADP before 25th February and 2-day ASP before 4th March, surface properties of roads and facades and the sampling methods of WVS and SW.

The wash-off concentrations of trace metals were identified in the following orders: Cd (2.28 ± 2.08 µg/l) < Pb (435.85 ± 412.61 µg/l) < Cu (0.93 ± 0.61 mg/l) < Zn (2.52 ± 2.30 mg/l). Accordingly, the wash-off concentrations revealed a considerable variation depending on the metal element, surface properties, antecedent environmental conditions, and sampling methods. The highest wash-off concentrations of Cd, Pb, Cu and Zn determined by SW (indicating a normal potential pollution) and WVS (indicating a maximum potential pollution) methods were 5.57 ± 2.36 µg/l, 6.08 ± 0.21 µg/l; 460.00 ± 112.80 µg/l, 965.00 ± 159.03 µg/l; 0.96 ± 0.20 mg/l, 2.00 ± 0.56 mg/l; and 3.45 ± 0.48 mg/l, 7.59 ± 1.46 mg/l, respectively. The highest concentrations determined by both methods of most of metals were measured on the road surface after ADP, except that the highest concentration determined by WVS for Pb was found on the facade surface after ADP.

The lowest wash-off concentrations of Cd, Pb, Cu, and Zn determined by SW method were 0.22 ± 0.19 µg/l, 64.30 ± 35.55 µg/l, 0.11 ± 0.07 mg/l, and 0.23 ± 0.13 mg/l, respectively. Those determined by WVS method were 1.46 ± 1.25 µg/l, 141.93 ± 81.77 µg/l, 0.35 ± 0.19 mg/l, and 0.88 ± 0.41 mg/l, respectively. The lowest concentration of Pb determined by the WVS method occurred on the road surface after ASP, while the lowest concentrations determined by both methods for the other metal were found on the facade after ASP. Cu concentration (1.22 ± 1.51) collected under the condition of facade/ADP/SW shows a large deviation, which would cause high uncertainty of the result.

Besides, Zn and Cu are typical anthropogenic pollutants [27,42].
The relatively higher content of both metals is primarily attributed to the corrosion of metal surface and vehicle abrasion in road traffic [17,43]. Pb was mainly contributed by gasoline-powered vehicles, but it was banned in Germany in 1996 [44]. Thus, the presence of Pb on the surface would come mostly from the past usage of leaded fuel [45,46] and partly from vehicle tires and brakes [47]. Finally, the low content of Cd accumulated on the surface might be contributed by atmospheric deposition [48].

In addition, the wash-off concentrations of TS showed the ranges from 5.23 to 10.49 g/l on road surface after ADP, 0.35–1.51 g/l on the facade surface after ADP, 2.40–5.69 g/l on the road surface after ASP, and 0.05–1.08 g/l on the facade surface after ASP.

3.2. Influence of antecedent environmental conditions on the contents of metal and TS

Fig. 2 illustrates the wash-off concentrations of Cd, Pb, Cu, and Zn determined by SW and WVS methods. It is obvious that the...
wash-off concentrations of all trace metals and TS on either road or low-elevated facade surface were significantly higher after ADP than those after ASP. The results suggest that the snowmelt in the antecedent period after a snowfall could considerably influence the accumulation of metals on the surface in the following rainfall event. The concentrations of Cd, Pb, Cu, Zn, and TS determined by the SW method in road surface runoff after ASP were 51%, 74%, 38%, 42%, and 54%, respectively. Compared with ADP, ASP showed a clearer impact on the metal content level in the runoff from the low-elevated facade surface. Their concentrations also determined by the SW method in the facade surface runoff after ASP were 80%, 83%, 91%, 79%, and 84%, respectively. The wash-off concentrations collected by the WVS method showed the same results.

In terms of the individual metal, Pb was the anthropogenic metal that received a strong impact from snow events. It is because the primary source of Pb, past usage of leaded fuel, was considerably more inactive than the pollution sources of other metals, e.g. atmospheric deposition for Cd and vehicle-related sources for Cu and Zn. It would be significantly influenced by the continued wash-off process from snowmelt. Compared with the content of metals, the snowmelt showed a relatively stronger influence on the wash-off concentration of TS as determined by the SW method. Generally, snowpack can absorb the atmospheric deposited particles and other pollutants [49]. During the snowmelt, the particulate matters will be coagulated with other particles and move slowly in the snowpack [50]. Therefore, the build-up rate of deposited solids on the surface after ASP will be slower than it after ADP. However, the snowmelt had a limited impact on the TS wash-off content determined by the WVS method. It suggests that the snowmelt had a stronger influence on the easily detached particles.

Although the result indicates that the snowmelt reduced the pollution in rainfall-induced surface runoff, the snowpack might contain a large load of pollutants [50,51]. The rain-on-snow can actually flush the pollutants from the snowpack and possibly cause even higher pollution load in the runoff. Therefore, the influence of snowmelt during rainfall events on pollutants in the runoff should be further evaluated.

3.3. Understanding the variance on the road and low-elevated facade surfaces

In comparison with the two analyzed surfaces under the same sampling conditions, generally, the concentrations of trace metals on the road were much higher than those on the facade (Fig. 2). Specifically, the content of Cd, Cu, and Zn washed off the road surfaces was commonly 45–780% higher than them off the facade surfaces. The amount of TS on the road surface was even 420–4500% higher than them on the facade surface. The gravity force and roughness property of the facade might be the reason since only the particles with strong adhesion capacity can be attached stably on the facade surface. Pb content in surface runoff determined by the SW method had comparable concentrations on the road and facade surface. However, the higher Pb content measured by the WVS method was washed off from the low-elevated facade surface than the road surface for both ADP and ASP. It might because a relatively higher Pb content was usually found from bricks made from mine tailings [30,52].

Besides, the concentration changes in trace metals and TS from the road to facade surfaces after ADP were consistent with the changes after ASP. It indicates that the pollutants on the low-elevated facade were closely related to the solids on the road surface. Therefore, trace metals and TS on the low-elevated facade surface should mainly originate from the re-suspended pollutants and solids from the surrounding area.

Fig. 3 illustrates the ratios of trace metal and TS concentrations in different conditions. It reveals the trace metal content absorbed on the unit gram of particles and all ratios on the facade surface were always higher than on the road surface under the same antecedent weather condition by both sampling methods. The ratios on the facade surface were 2–13 times higher than them on the road surface. It suggests that each gram of particles on the facade surface contained more content of trace metals. It also showed the relatively higher concentration ratios when using the SW sampling method than the WVS method, which might be attributed to the fact that more particles with less trace metal content were washed off when using the WVS method.

3.4. Sensitivity analysis based on Bayesian network

The sensitivity analysis was derived from the entropy reduction with the Bayesian network. The entropy reduction was shown in Fig. 4. The antecedent environmental conditions of ADP and ASP, surface properties of roads and facades, sampling methods of WVS and SW were the main variables exerting influences on the concentrations of trace metals.

With regard to Cd and Zn, they had similar results of classification. The primary influencing variable on their wash-off concentrations was the surface properties. The antecedent environmental conditions and the sampling methods had a minor influence on the wash-off concentrations. It showed the same results displayed in Fig. 2 that the concentration reduction rate from the road to the facade surface (averagely 72% for Cd; 74% for Zn) was obviously higher than the antecedent environmental conditions (from ADP to ASP: 53%; 61%) and the sampling methods (from WVS to SW: 39%; 52%) when only one influencing variable was changed at a time.

In terms of Pb and Cu, both concentrations predominantly depended on the antecedent environmental conditions. The concentration reduction rate from ADP to ASP was 81% for Pb and 66% for Cu, which was highest among all variables. For Pb, the application of different sampling methods had a relatively stronger impact on surface conditions. It was agreed with the finding that the Pb concentrations in WVS samples were particularly higher on facade than road surface. In contrast, the surface conditions for Cu showed the comparable impact to the antecedent environmental conditions. It might be attributed to the high uncertainty under the condition of facade/ADP/SW as shown in Fig. 2.

4. Conclusions

The main focuses of this study are to determine the influence of antecedent weather conditions on the metals' build-up process and their further levels in the surface runoff. The result shows that the snow-melting event had a considerable influence on the wash-off concentrations of the trace metals on both road and facade surfaces. The wash-off concentrations of Cd, Cu, and Zn on the facade surfaces were lower than them on the road surface. More metals contents could be measured using the WVS than SW, thus WVS could indicate a maximum pollution potential in the surface runoff. The results of sensitivity analysis based on the Bayesian network show that the concentrations of Cd and Zn were mainly dependent on the surface properties, while the antecedent environmental conditions strongly influenced the concentrations of Pb and Cu. The sampling methods had a minor influence on the wash-off concentrations.
The data provided herein could assist a better understanding of the wash-off process and an improving modeling for the pollutant migration in surface runoff by considering the different surfaces. The influences of snowmelt and rainfall events with a longer antecedent period should be further evaluated.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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