Sorption behavior of heavy metal species by soakaway sediment receiving urban road runoff from residential and heavily trafficked areas

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ABSTRACT

Groundwater contamination by heavy metals from infiltration facilities receiving road runoff is of potential concern. In this study, sorption tests were conducted to evaluate the influence of the water quality of road runoff, especially dissolved organic matter (DOM), on the sorption of heavy metal species by soakaway sediment. Sequential batch tests were conducted to assess metal sorption by the soakaway sediment receiving road runoff from residential and heavily trafficked areas. Ni was adsorbed by the sediment, indicating that soakaway sediments function to prevent groundwater contamination by Ni. In contrast, Zn was released from the soakaway sediment in sorption tests using heavily trafficked road dust leachates. Ni, Cu, Zn, and dissolved organic carbon concentrations were higher in soakaway sediment leachates obtained by sorption tests using heavily trafficked road dust leachates than those using residential road dust leachates, suggesting traffic activities contaminate these pollutants. A large portion of Zn, released from the soakaway sediment, existed as stable complexes. DOM in road runoff possibly enhances the release of Zn from the sediments within infiltration facilities and might cause groundwater contamination.

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1. Introduction

Urban road runoff is contaminated by non-point pollutants such as heavy metals derived from traffic activities and has been regarded as an important pathway to water environments [1–6]. Effective strategies are needed to prevent water environment pollution through urban road runoff. Among possible management systems, infiltration of road runoff is a promising option to control the pollutants.

Infiltration facilities receiving urban road runoff were originally designed to reduce runoff peak flow and replenish groundwater [7,8]; however, they are also expected to trap non-point pollutants such as heavy metals and thus to reduce their discharges to water environments [9,10]. For example, Mikkelsen et al. revealed that the content of non-point-pollutants such as heavy metals were specifically higher in surface of soils in the infiltration systems, suggesting that soils in infiltration facilities acted as sorbent of pollutants in road runoff [9]. Several researchers also supported this accumulation of non-point pollutants onto solids within or below infiltration facilities [11–14]. Some researchers, however, have pointed out that

groundwater contamination by heavy metals from infiltration facilities is also of potential concern [15–17]. This potential concern is a major barrier to the use of infiltration facilities for road runoff. It is required to assess the sorption (adsorption and desorption) of heavy metals by sediments within infiltration facilities receiving urban road runoff.

Toxicities and bioavailabilities depend on heavy metal species, e.g. free ion and labile complexes are more toxic and bioavailable to aqueous organisms and plants [18–21]. The information on speciation of heavy metals is useful because removals of heavy metals in runoff by plant uptake are sometimes expected [22]. Besides, transport of heavy metals is also related to their speciation. In particular, complexation with dissolved organic matter (DOM) often governs the solubility and mobility of heavy metals [23–26]. For example, Walsh and O'Halloran found that stable organic acid complexes of Cr(III) are likely to be relatively persistent in the estuary [24]. Organic complexes of heavy metals were often found to be dominant in groundwater and landfill leachates [23,27–29]. Therefore, the sorption of heavy metal species in infiltration facilities might be affected by the water quality characteristics of road runoff (e.g. DOM). We previously evaluated the speciation of heavy metals and their behavior in infiltration facilities, highlighting that Cu dominantly existed as organic complexes and that Zn was released from sediment in the soakaway [30]. However, there were limited

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discussions on relationship between the sorption behavior of heavy metal species and the water quality of road runoff. To demonstrate the role of the water quality characteristics of road runoff, especially DOM, for the transport of heavy metal species in infiltration facilities, it is now required to investigate the relationship by using several types of road runoff samples with different characteristics.

The objective of this study is to understand the sorption behavior of heavy metal species by sediments in infiltration facilities receiving urban road runoff. We investigated the influence of the water quality of road runoff, especially DOM, on the sorption of heavy metal species by the soakaway sediment. In this study, sequential batch tests, which mimicked the sorption process, were conducted by adding several types of artificial road runoff water (road dust leachates) to the soakaway sediment to obtain artificial percolating water from the soakaway sediment (soakaway sediment leachates).

2. Materials and methods

2.1. Samples

Soakaway sediment was collected with a polyvinyl chloride tube (corer) from bottom to top from a residential area with an infiltration-type sewage system in Tokyo that was constructed in the 1980s [7]. The major components of a sediment in the same soakaway collected on a different day, determined by electron probe microanalyzer, were reported in [14], showing that the sediment was comprised of particles whose major components were [O, Al, Si], [O, Si] or [O, Mg, Si]. More than 90 g of road dust was also collected using a vacuum cleaner (Hitachi CV-100S6) from each road gutter surface in the residential areas and heavily trafficked areas (traffic volumes: 19,848–36,666 vehicles/day [31]) in Tokyo. After the road dust sampling was finished, the vacuum cleaner was cleaned by another vacuum cleaner to minimize any risk of cross-contamination. The road dust and sediment were air-dried at room temperature and homogenized after being sieved through 2 mm mesh to remove larger particles and stones. Table 1 shows the sampling conditions, the heavy metal contents and the ignition loss. The antecedent rainfall and dry weather period data were obtained from a meteorological database [32]. Ni, Cu and Zn were targeted because they were derived from traffic activities [33]. We previously used the samples of the soakaway sediment and the heavily trafficked road dust #1 for the sorption tests [30,34]. The heavy metal contents and the ignition loss were reported by Murakami et al. [30,33] and Fujita et al. [34]. To assess the pollution status of the soakaway sediment used in this study, Fig. 1 compares the heavy metal contents and ignition loss in soakaway sediments between this study and our previous study [33], which revealed their spatial distributions in surface sediments (≤10 cm depth) in 16 soakaways in the same residential area (N35°44′34–37″ E139°35′42–53″). The soakaway sediment used in this study contained a relatively high level of heavy metal contents and organic matter, probably due to significant adsorption, which resulted from infiltration of urban runoff through the soakaway during long-term use.

2.2. Sequential batch tests

To mimic the sorption behavior in the soakaway, the sequential batch tests were conducted following the previous study [30]. Fig. 2 shows a procedure of the sequential batch tests. These tests allow the setup of a reproducible, reliable and constraint-free experimental scheme. First, road dust leachates were prepared to mimic road runoff from residential and heavily trafficked areas. Deionized water (DW) and the road dust (residential road dust #1–4 and heavily trafficked road dust #1–4) were mixed at an L/S ratio of

Table 1
Sampling conditions, heavy metal contents and ignition loss in road dust and soakaway sediment

	Residential road dust				Heavily trafficked road dust				Soakaway sediment
	#1	#2	#3	#4	#1[a]	#2	#3	#4	
Sampling conditions									
Sampling date	17 September 2004	17 September 2004	17 September 2004	17 September 2004	5–6 November 2004	5 November 2004	6 November 2004	18 November 2004	27 June 2005
Coordinates (WDS-84)	N35°44′36″ E139°35′53″	N35°44′35″ E139°35′51″	N35°44′35″ E139°35′53″	N35°44′37″ E139°35′44″	N35°43′07″ E139°44′37″ N35°43′07″ E139°45′51″	N35°42′29″ E139°46′04″	N35°42′58″ E139°44′21″	N35°42′46″ E139°45′34″	N35°44′37″ E139°35′44″
Traffic volume (vehicles/day)[b]	–	–	–	–	17030–22138	19848	34712	36666	–
Antecedent rainfall (mm)[c]	4.0	4.0	4.0	4.0	17.5	17.5	17.5	17.5	22.0
Antecedent dry weather periods (h)[c]	160	160	160	160	103–125	99	123	171	119
Heavy metal contents and ignition loss[d]									
Ni (mg/kg)	49	41	42	67	82	97	79	96	69
Cu (mg/kg)	100	90	170	130	510	990	730	600	340
Zn (mg/kg)	1200	450	1800	1300	1300	1900	1400	1500	1500
Ignition loss (%)	6.1	6.3	12	10	9.3	13	11	14	20

[a] The road dust collected from two lines was mixed at a dry weight ratio of 1:1.
[b] These data are taken from Japan Society of Traffic Engineers [31].
[c] These data are taken from a meteorological database [32], at the closest monitoring station. Light rain (≤3 mm) is excluded.
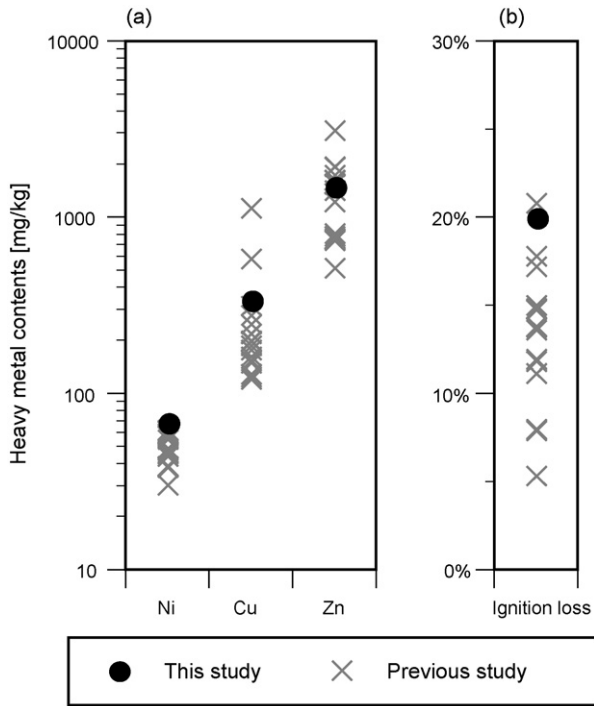[d] Heavy metal contents and ignition loss are given in [30,33,34].

Fig. 1. Comparison of heavy metal contents and ignition loss between this study and previous study ($n = 16$) [33]: (a) heavy metals and (b) ignition loss.
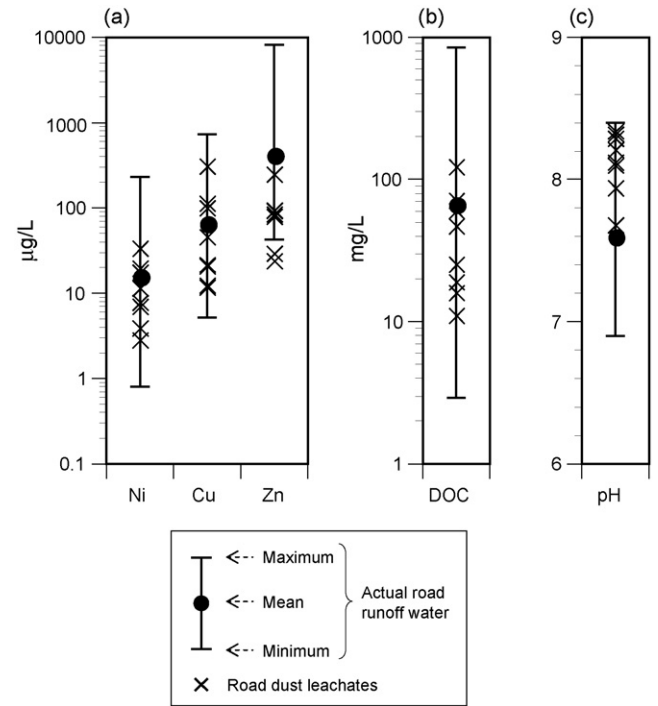

Fig. 3. Comparison of chemical parameters in dissolved phases between road dust leachates and actual road runoff water: (a) heavy metals, (b) DOC, and (c) pH. The following values for actual road runoff are obtained from the literature: heavy metals and DOC, n (total number of events) = 62 [6]; pH, $n = 6$ [1].
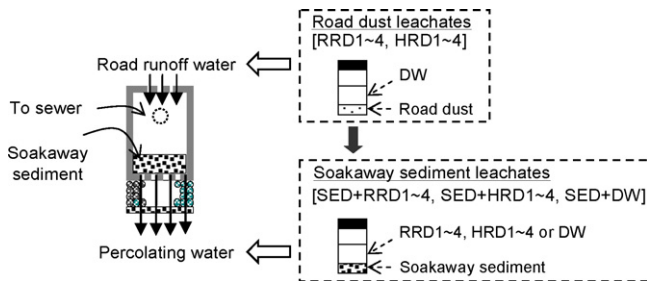

Fig. 2. Conceptual diagram of sequential batch tests (DW: deionized water).

10 L/kg dry material and shaken for 16 h at a speed of 200 rpm at room temperature in the dark. Deionized water were utilized as a solvent because there were no large differences in pH, dissolved organic carbon (DOC), heavy metal, major cation and anion concentrations between deionized water and 1 mM HNO_3 as the solvent to create road dust leachates [30]. The road dust leachates were obtained by centrifugation ($1600 \times g$, 20 min) and filtration of the supernatant with a PTFE membrane filter (Advantec, H050A, pore size: 0.5 μm): i.e. residential road dust leachates, RRD1∼4; heavily trafficked road dust leachates, HRD1∼4. Then, to simulate the sorption phenomenon in soakaways, sorption tests were conducted by adding the road dust leachate or DW to the soakaway sediment and shaking as described above. The soakaway sediment leachates
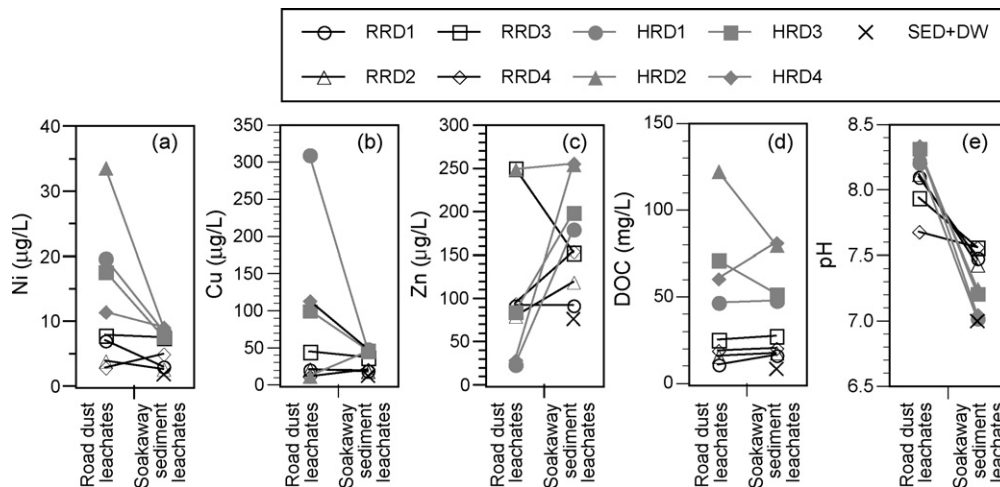

Fig. 4. Comparison of heavy metal concentrations, DOC concentrations, and pH between road dust and soakaway sediment leachates: (a) Ni, (b) Cu, (c) Zn, (d) DOC, and (e) pH. Data of HRD1, SED + HRD1 and SED + DW are given in [30,34].

(SED + RRD1~4, SED + HRD1~4, and SED + DW) were obtained after centrifugation and filtration.

2.3. Chemical analyses

DOC, pH and heavy metals (Ni, Cu and Zn) were measured in all of the leachates. DOC was determined by a TOC analyzer (Shimadzu TOC-V CSH). The pH was measured with electrodes. One split of filtered sample was microwave-digested (170 °C, 10 min) with concentrated nitric acid (a sample: concentrated nitric acid = 9:1 (v/v)) for quantification of the total dissolved metals in the leachates. Free ion and labile complexes of heavy metals were determined with the chelating resin disk cartridge (Empore, iminodiacetate functionalized poly[styrenedivinylbenzene]), as described in Murakami et al. [30]. In brief, 5 mL of 3.0 M nitric acid, 10 mL of DW and 5 mL of 100 mM ammonium acetate (pH 5.5) were sequentially passed through the cartridge. Then, one split of filtered sample was passed through the cartridge, and 5 mL of DW was passed through the cartridge as a rinse. The trapped heavy metals were eluted with total 3 mL of 3.0 M nitric acid. Internal standards (Sc and Y) were added to all the samples. All the samples were acidified at pH < 1 with nitric acid. The free ion and labile complexes, and total dissolved fractions of heavy metals in the leachates were then measured by inductively coupled plasma-mass spectrometry (ICP-MS; Yokogawa HP 4500). The internal standards with the corresponding analytes assigned for quantification were as follows: ^{45}Sc for ^{60}Ni; ^{89}Y for ^{63}Cu and ^{64}Zn. The stable complexes were determined from the difference between the total dissolved and free ion and labile complex fractions of heavy metals. The pH, DOC, and heavy metal concentrations in HRD1, SED + HRD1 and SED + DW were reported by Murakami et al. [30], and Fujita et al. [34].

2.4. Statistical analysis

A two-way analysis of variance (ANOVA) of mixed design was used to test the difference of heavy metal and DOC concentrations: i.e. between-groups design, traffic activities (RRD vs HRD and SED + RRD vs SED + HRD); repeated-measures design, sorption (RRD vs SED + RRD and HRD vs SED + HRD). Logarithmic data of heavy metal and DOC concentrations were applied to the test. SPSS 14.0J was used for statistical analyses.

3. Results and discussion

3.1. Comparison of chemical parameters between road dust leachates and actual road runoff water

Road dust leachates in this study were prepared by mixing deionized water with road dust for 16 h, which might be different from actual conditions of road runoff. This needs to demonstrate whether the prepared road dust leachates are similar to actual road runoff. We previously confirmed that the heavy metal, DOC concentrations and pH in road dust leachates were within the range of those in dissolved phase of actual road runoff water in literatures, suggesting that the road dust leachates could be regarded as valid and representative to mimic road runoff water [30,35]. Again, we demonstrated the validity of road dust leachates prepared in this study. Fig. 3 compares chemical parameters (heavy metals, DOC and pH) of road dust leachates with those in dissolved phase of actual road runoff water reported in the literatures [1,6]. The values of chemical parameters in the road dust leachates were within the ranges reported in the literatures for the actual road runoff water, except Zn in two road dust leachate samples. Therefore, the road
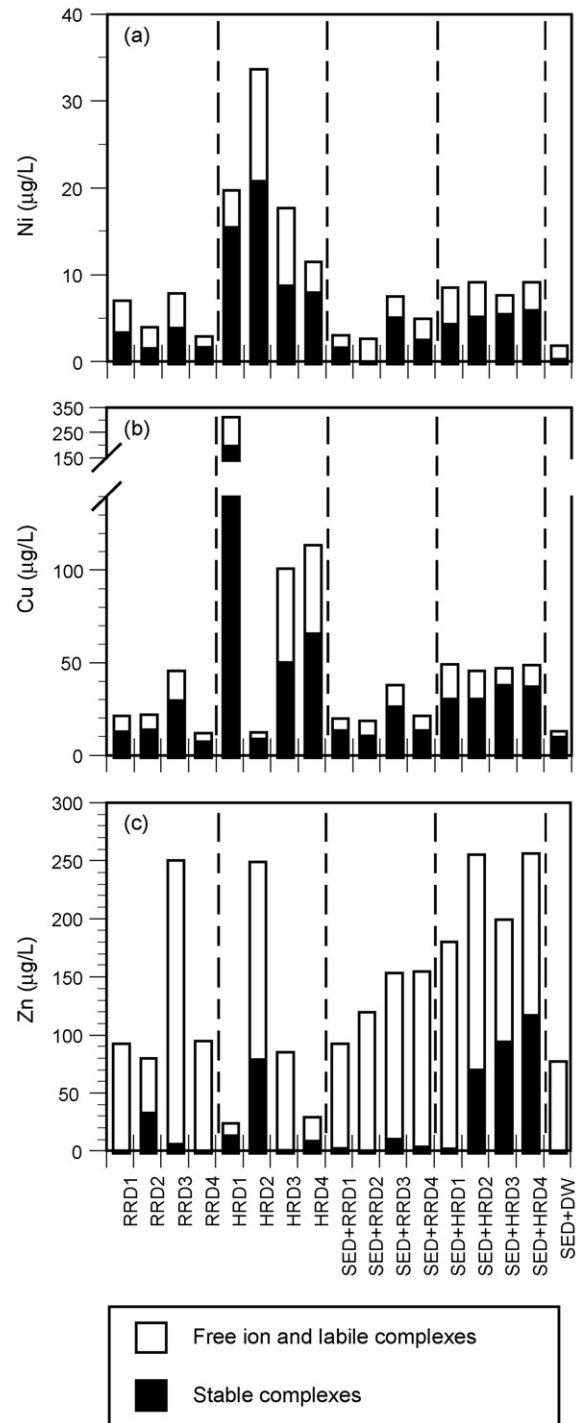

Fig. 5. Speciation of heavy metals determined from chelating resin fractions: (a) Ni, (b) Cu, and (c) Zn. Data of HRD1, SED + HRD1 and SED + DW are given in [30,34].

dust leachates, newly prepared in this study, could be regarded as valid.

3.2. Sorption of heavy metals in road runoff with different characteristics by soakaway sediment

Fig. 4(a–d) compares total dissolved heavy metal and DOC concentrations between road dust and soakaway sediment leachates. The pH is also given in Fig. 4(e) as the supporting information
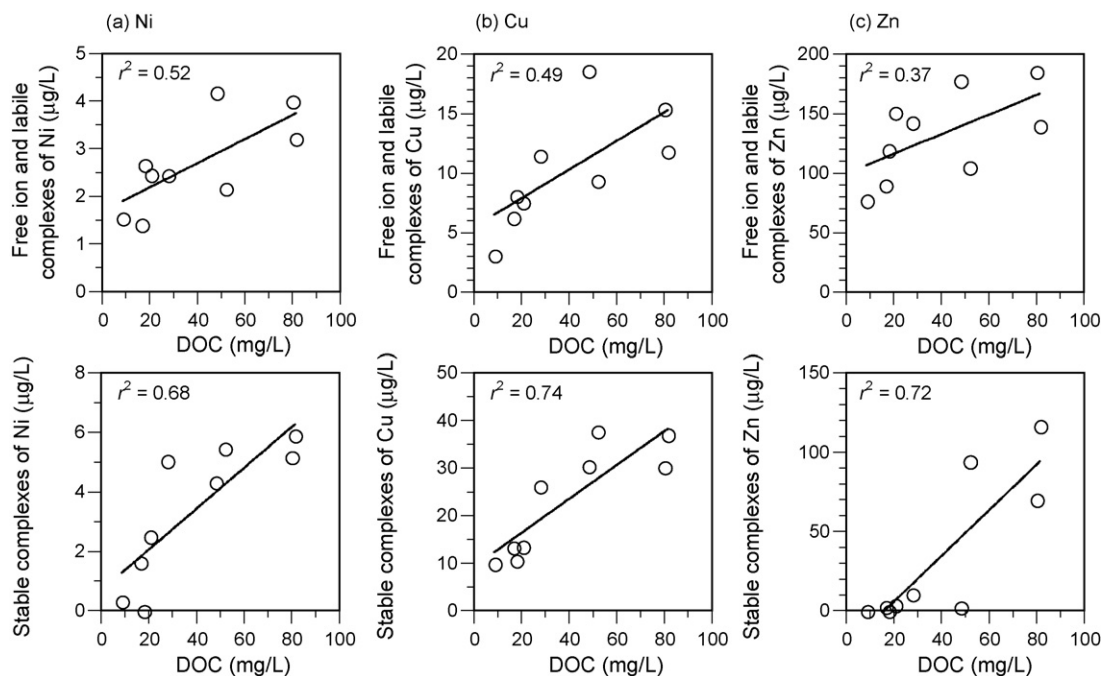
Fig. 6. Correlation between DOC and heavy metal species in soakaway sediment leachates: (a) Ni, (b) Cu, and (c) Zn.

on the sorption behavior. Ni concentrations in soakaway sediment leachates (2.7–9.2 μg/L) were significantly lower than those in road dust leachates (2.9–34 μg/L) ($F(1,6) = 7.22$, P (probability) < 0.05), indicating that the soakaway sediment adsorbed Ni in road runoff. This result is worthy of special mention that the soakaway sediment with 2-decade operations was useful to prevent groundwater contamination by Ni, because Ni is the one of potential groundwater pollutants due to the slight removals by loamy soils [35]. In contrast, there were insignificant differences in Cu and DOC concentrations between road dust and soakaway sediment leachates (Cu: road dust leachates, 12–310 μg/L, soakaway sediment leachates, 19–49 μg/L; DOC: road dust leachates, 11–120 mg/L, soakaway sediment leachates, 17–81 mg/L) (Cu: $F(1,6) = 0.53$, $P > 0.20$; DOC: $F(1,6) = 0.10$, $P > 0.20$), suggesting the soakaway sediment was not useful to remove these pollutants in road runoff. This figure also compares the chemical parameters in leachates prepared from residential and heavily trafficked road dust. Ni, Cu, and DOC concentrations were significantly higher in heavily trafficked samples (HRD and SED + HRD) (Ni, 7.6–34 μg/L; Cu, 12–310 μg/L; DOC, 47–120 mg/L) than in residential samples (RRD and SED + RRD) (Ni, 2.7–7.9 μg/L; Cu, 12–46 μg/L; DOC, 11–28 mg/L) (Ni: $F(1,6) = 21.77$, $P < 0.05$; Cu: $F(1,6) = 6.22$, $P < 0.05$; DOC: $F(1,6) = 37.45$, $P < 0.05$), suggesting that heavy traffic activities contaminated these pollutants.

On the other hand, Zn showed a significant interaction between traffic activities and sorption ($F(1,6) = 4.42$, $P = 0.08$), as shown in Fig. 4(c). Unlike Ni, Cu and DOC, Zn concentrations increased significantly after mixing the heavily trafficked road dust leachates and the soakaway sediment (HRD, 24–250 μg/L; SED + HRD, 180–260 μg/L) ($F(1,6) = 10.41$, $P < 0.05$). This was consistent with the previous finding that in soakaway sediments, Zn was more highly mobile than Ni and Cu [36]. Besides, Zn concentrations were significantly higher in SED + HRD (180–260 μg/L) than in SED + RRD (92–150 μg/L) ($F(1,6) = 13.18$, $P < 0.05$), while Zn concentrations in HRD (24–250 μg/L) were comparable to or lower than those in RRD (80–250 μg/L) ($F(1,6) = 1.06$, $P > 0.20$). These results indicated that heavily trafficked road runoff enhanced the release of Zn from the soakaway sediment and might cause groundwater

contamination, even though Zn concentrations in heavily trafficked road runoff were comparable to or lower than those in residential road runoff. This highlighted possible influence of the water quality of road runoff on sorption behavior of Zn within infiltration facilities.

3.3. Relation between sorption behavior of heavy metal species and DOM

Fig. 5 shows speciation of heavy metals determined from chelating resin fractions. Ni and Cu dominantly existed as stable complexes in both road dust and soakaway sediment leachates. In contrast, Zn predominantly existed as free ion and labile complexes in leachates except SED + HRD. Concentrations of labile complexes are similar with those of inorganic complexes, whereas stable complexes are most likely organic complexes [35,37]. Hence, it indicated that complexation with DOM played more important roles in speciation of Cu and Ni than Zn. This was consistent with the previous findings that complexation with DOM was more significant for Cu than for Zn, although consistent tendencies were not found for Ni [28,38]. However, in SED + HRD leachates (except SED + HRD1), stable complexes of Zn were also present. Stable complexes of Zn were not present in SED + HRD1, probably because HRD1 had lower organic matter contents (Table 1) and DOC concentration (Fig. 4(d)) than other HRD samples. The relatively low traffic volume of HRD1 (Table 1) might be attributed to this difference. The abundance of stable complexes of Zn in SED + HRD leachates (except SED + HRD1) suggested that the release of Zn from soakaway sediments may be related to complexation with DOM. This was supported by a strong correlation between DOC and stable Zn concentrations in soakaway sediment leachates ($r^2 = 0.72$) but a weak correlation between DOC and free ion and labile Zn concentrations ($r^2 = 0.37$), as shown in Fig. 6. DOM in road runoff possibly acts to facilitate the release of Zn from the sediments within infiltration facilities. Also, there were stronger relationships between DOC and stable Ni or Cu concentrations than between DOC and their free and labile concentrations. This indicated that DOM functioned to retain Ni

and Cu in percolating water from the soakaway sediment. This is consistent with the past findings that solubility and mobility of heavy metals are often controlled by complexation with DOM [23–26]. DOM in road runoff possibly suppress adsorption of heavy metals and sometimes enhances the release of them from the soakaway sediment and might cause groundwater contamination.

3.4. Recommendations

Overall, the concentrations of all the heavy metals (Ni, Cu and Zn) were higher in SED + HRD than SED + RRD, even though the concentrations in heavily trafficked road runoff were comparable to or lower than those in residential road runoff like the case of Zn. DOM in road dust leachates probably acted to release stable complexes of heavy metals from the soakaway sediment. This is of particular concern because stable heavy metals resist removals by natural organic media (e.g. plant [22], and hardwood mulch [39]) and possibly artificial adsorber media [26,40]. Therefore, these results indicated that controlling sources of not only heavy metals but organic matter would help to prevent groundwater contamination by heavy metals from infiltration facilities.

4. Conclusions

In this study, sequential batch tests were conducted to evaluate the influence of the water quality of road runoff, especially DOM, on the sorption behavior of heavy metal species by soakaway sediment. We draw the following conclusions:

- The soakaway sediment adsorbed Ni in road runoff, whereas Zn was desorbed from the soakaway sediment in sorption tests using heavily trafficked road dust leachates. Ni, Cu, Zn, and DOC concentrations were higher in soakaway sediment leachates obtained by sorption tests using heavily trafficked road dust leachates than those using residential road dust leachates.
- Ni and Cu dominantly existed as stable complexes. Stable complexes of Zn were also present after mixing the heavily trafficked road dust leachates and the soakaway sediment.
- Heavily trafficked road dust leachates facilitated to release stable complexes of Zn from the soakaway sediment. DOM in road runoff possibly enhances the release of Zn from soakaway sediments.

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