



## LEACHING BEHAVIOR OF NUTRIENTS AND METALS OF GREEN ROOFS: LABORATORY AND FIELD STUDIES

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**Abstract:** Despite the benefits of green roofs in managing urban stormwater, chemical leaching especially at their initial stage can be problematic. Although chemical leaching from green roofs has been acknowledged, to date research in this aspect is still limited. In this paper, both laboratory and field experiments were conducted to investigate and characterize the temporal evolution of nutrient and metal leaching. In the laboratory study, three cells were constructed with three types of growing media and their chemical leaching was monitored under simulated rain events; while the chemical leaching from a full-scale green roof on the Calgary's Municipal Building was monitored in natural events and compared to a nearby conventional roof and the cells. The water samples (collected from the cells, the green roof, and the conventional roof) were analysed for nutrients and metals. Statistical approaches were adopted to investigate their leaching behaviour and to assess the primary influential factor which could be used to capture their temporal evolution. The observations showed that both the laboratory cells and full-scale green roof acted as a source of these pollutants (except two metals in the full-scale green roof). The laboratory observations also showed that nutrient leaching in general decreases with the cumulative inflow and the leaching response to the cumulative inflow appeared to be different for different types of media. Similar results were also observed from the full-scale green roof, especially for  $\text{PO}_4^{3-}$ , and TP. The results would help in enhancing the design, application and modelling of green roofs.

### 1 INTRODUCTION

In urban settings, population growth and consequent urbanization have posed significant adverse impacts on urban ecosystem due to the sharpened stormwater runoff hydrograph and degraded stormwater runoff quality. Thus, it is essential to eliminate or attenuate such adverse effects to maintain sustainable urban environment. As the conventional stormwater management technologies have shown their constraints (e.g. space consuming, low efficiency in removing many pollutants, and high cost in construction, maintenance, and rehabilitation) in managing urban stormwater, low impact development technologies (LIDs) as an alternative to the conventional technologies have recently attracted attention. The green roof is one of typical LIDs and has been demonstrated to have dual benefits in managing urban stormwater (in both quantity and quality) (Mentens *et al.*, 2006; Li and Babcock 2014). However in contrast to the concept green roofs could act as a source rather than a sink of various pollutants (especially nutrients) in their early age, as the growing media (GM) of green roofs are often supplemented with compost or fertilizer to sustain vegetation growth (Bliss *et al.*, 2009). In addition, metal leaching is common and its management is challenging (Alsup *et al.*, 2011). The existing body of knowledge demonstrates that the behaviour of a green roof (source or sink) is dependent on GM properties, roof design and age, vegetation type, and the use of

fertilizer or compost (Emilsson *et al.*, 2012). However green roofs could be sinks eventually after exchangeable pollutants are exhausted (Alsup *et al.*, 2011).

Although chemical leaching from green roofs has been well acknowledged, to date very limited research has been conducted to link their chemical leaching behavior to potential influential factors including design and hydrological variables qualitatively and quantitatively. Furthermore, to the authors' best knowledge, no research so far has focused on the temporal behaviour of chemical leaching and the investigation of the translatability of knowledge from laboratory to field in this aspect. Therefore, this paper observed chemical leaching of green roofs through both laboratory and field experiments aiming to quantify the effects of the influential factors quantitatively. The results would help in enhancing green roof design, application and modelling to reduce chemical leaching.

## 2 MATERIAL AND METHODS

### 2.1 Laboratory Experiments

The laboratory experiments were conducted using simulated events to investigate the effects of design variables (GM type) and hydrological variables (rainfall intensity and antecedent moisture condition (AMC)) on leaching of nutrients and metals. Three laboratory cells were constructed using three types of GM including ZinCoblend-SI (GM1), Eagle Lake rooftop media blend (GM2), and SOPRAFLOR I (GM3), which are available locally or applied in the City of Calgary. The three cells were exposed to three rainfall magnitudes (2-, 5- and 10-yr storms) and three degrees of AMC (dry (<20%), normal (20-30%), and wet (>30%)). Each cell (Figure 1a) has a surface area of 0.57m x 0.41m, a depth of 150 mm, and a 1.5cm diameter drainage hole at the bottom. The laboratory cells have same structure as the full-scale extensive green roof (GR) investigated in the paper. The cells consist of a layer of 150-mm GM vegetated with sedum, a filter sheet, a drainage layer (FD 25-E), and protection mat (SSM 45). Sedum are locally available and most commonly adopted due to their tolerance to extreme weather condition (e.g., extreme temperature and high wind speed) and the limited water consumption criteria (VanWoert *et al.*, 2005; Berrett *et al.*, 2014). The slope of the cells was fixed at 1% to mimic the slope of the GR. The threshold values of AMC were determined based on average GM moisture (using 2015-2016 data) measured from the GR. The rainfall duration was fixed at 1-hour in all experiment runs as it has been used to examine the operation of source control practices such as green roofs in the City of Calgary (The City of Calgary, 2011). Thus, the rainfall intensities, which is uniform over the duration, corresponding to 2-, 5-, and 10-yr storms are 14.09, 20.27, and 24.31mm/hr, respectively.

Due to varying these design and hydrological variables, a total of 27 experiments were conducted. In each experiment run, deionized water was applied to the cells, thus no additional pollutants were added to the cells. A composite sample from each experiment run was collected and analyzed for nutrients including nitrate ( $\text{NO}_3^-$ ), ammonia ( $\text{NH}_4^+$ ), total nitrogen (TN), orthophosphate ( $\text{PO}_4^{3-}$ ), and total phosphorus (TP), and metals including zinc (Zn), copper (Cu), and lead (Pb). Their Event Mean Concentrations (EMCs) were reported. In addition, the GMs were analyzed for their initial contents of nutrients and metals using the water extraction method (Hurley *et al.*, 2017), which is a modified version of the field leach test of U. S. Geological Survey. All water samples were analyzed for nutrients using Hach methods and for metals using 7900 ICP-MS at Calgary's laboratory in the Bonnybrook Wastewater Treatment Plant.

### 2.2 Field Experiments

A 335-m<sup>2</sup> extensive GR (Figure 1b) and a 335-m<sup>2</sup> conventional roof as the reference roof (RR), which are situated on the 5<sup>th</sup> and 4<sup>th</sup> floor of Calgary' Municipal Building in the downtown core, have been monitored since July 2015. Note that the 50% of GR surface is covered by gravels and hard roof. The GR was constructed using GM1 and its construction was completed in the summer of 2014. The GR and the RR were monitored in natural events using automated flow monitoring (using magmeter) and water sample collection systems. The automatic water samplers collected discrete samples (maximum 24 samples) in each event. A total of nine species of vegetation are randomly distributed in 85 quadrats of the GR. Irrigation was applied to during the vegetation establishment periods. Soil moisture sensors were deployed at five locations and two depths (top and 5 cm deep) at each location to record moisture of GM. The water samples

were analyzed for nutrients and metals. During the study period, eight events (five events in 2015 and three events in 2017) were monitored for the GR and the RR and the EMCs of nutrients and metals were calculated and used in the following analysis.

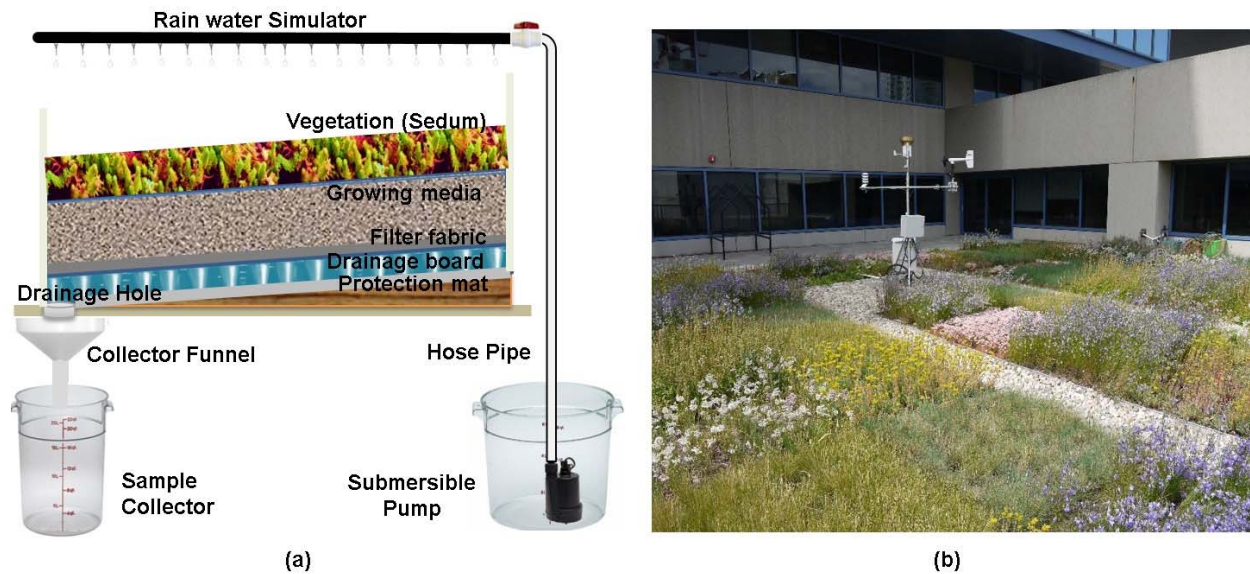


Figure 1. (a) The diagram of laboratory cells and (b) the full-scale green roof

### 2.3 Data Analysis

Considering the small sample size of the datasets, nonparametric Kruskal Wallis test was applied to compare the medians of the datasets. To investigate the sensitivity of the EMCs to the influential factor (cumulative inflow), the linear regression approach was employed. The significance of the regression slopes was tested using the t-test. In addition, the analysis of covariance (ANCOVA) test followed by the multiple comparison was applied to compare the slopes of the regression lines, which were detected to be significant, among or between the GMs for each water quality parameter. In this paper, a significant level of 10%, which is also often used in hydrological studies (e.g., Aziz and Burn, 2006; Hamed, 2008), was used.

## 3 RESULTS AND DISCUSSION

### 3.1 Initial Chemical Properties of the Growing Media

GM1 is high quality recycled materials and minerals and enhanced with compost; GM2 is the mixture of peat moss, fir bark fines, compost, sand, pumice and perlite; while GM3 composes of pumice, sand, vegetable compost, perlite, and blond peat mixed with high porous mineral aggregate. The initial chemical contents of the three GMs are presented in Table 1.

As illustrated in Table 1, the chemical properties in terms of the nutrient and metal contents are initially different among the GMs. The initial contents of TN and all metals are highest in the GM2; while the TP content is highest in the GM3. Similarly, a study conducted by the Toronto and Region Conservation Authority (TRCA) (2006) found large variations in both nutrient and metal contents among 11 commercially available GMs, for example TP and TKN (a component of TN) in the ranges of 239 - 1469 mg/kg and 191 - 3324 mg/kg, respectively; Zn, Cu, and Pb in the ranges of 25 - 133 mg/kg, 5 - 49 mg/kg, and 3 - 26 mg/kg, respectively. The initial TP content of the three GMs studied in the paper is below the range reported by TRCA; whereas TN content is within the range reported by TRCA. However, the contents of metals of the GMs (except GM3) are much higher than those reported by TRCA.

Table 1. Initial nutrient and metal contents of the three types of growing media.

Chemical content (mg/kg)	Type of GM		
	GM1	GM2	GM3
NO <sub>3</sub> <sup>-</sup>	110.00	444.87	147.93
NH <sub>4</sub> <sup>+</sup>	41.27	39.27	16.53
TN	239.40	819.00	259.80
PO <sub>4</sub> <sup>3-</sup>	109.33	88.87	168.87
TP	130.80	96.80	190.60
Zn	412.00	648.00	14.00
Cu	294.00	488.00	22.00
Pb	318.00	480.00	12.00

### 3.2 The Degree of Chemical Leaching in the Experimental Study

In a previous study conducted by the authors (Akther *et al.*, 2018), the GM type was identified as the most prominent influential factor of chemical leaching among the investigated design and hydrological variables including the thickness of GM, rainfall magnitude, and antecedent moisture condition. Therefore, in this paper the EMCs of the investigated water quality parameters were pooled and analyzed for individual GM type. Several descriptive statistics of the EMCs for each GM type are presented in Table 2. In addition, the results from the comparison of the medians of the EMCs among the three GMs using the Kruskal Wallis test are also included in Table 2. The presence of all pollutant constituents in outflow demonstrates that all the cells act as the sources of these pollutants. In addition, significant differences in the medians of the EMCs among three or between two GMs were detected in all nutrients and Zn in the Kruskal Wallis test.

As shown in Table 2, the medians/means and standard deviations of the EMCs of TN, NO<sub>3</sub><sup>-</sup>, and Zn of GM2 are obviously higher than those of GM1 and GM3; while the highest medians/means and standard deviations of the EMCs of TP and PO<sub>4</sub><sup>3-</sup> were observed in GM3. In addition, it was found that the medians of the EMCs of TP and PO<sub>4</sub><sup>3-</sup> are statistically significantly higher in GM3 than GM1 and/or GM2; while the medians of the EMCs of TN and NO<sub>3</sub><sup>-</sup> were detected to be statistically significantly higher in GM2 than in GM1 and GM3 in the Kruskal Wallis test. The median of the EMCs of NH<sub>4</sub><sup>+</sup> was found to be significantly lower in GM2 than in GM1 and GM3. Compared to NO<sub>3</sub><sup>-</sup>, the degree of the leaching of NH<sub>4</sub><sup>+</sup> is much lower. The results in general support that the GM of higher initial contents of nutrients (TP, PO<sub>4</sub><sup>3-</sup> and TN) leaches more nutrients and thus the degree of leaching of these constituents is associated with their initial contents in GM. However similar results were not observed for metals. The initial metal contents are highest in GM2. Although the highest medians of the EMCs of the metals were measured from GM2, only the median of the EMCs of Zn was found to be statistically significantly higher than GM1 and GM3. The medians of the EMCs of Cu and Pb are statistically equivalent among the GMs, however their initial contents of GM3 are obviously much lower than those of GM1 and GM2. These results might imply that the leaching of metals (especially Cu and Pb) is less dependent on their initial contents of GM compared to nutrients.

### 3.3 Characteristics of Chemical Leaching in the Experimental Study

The previous study by Akther *et al.* (2018) concluded that the nutrient leaching is primarily explained by GM type not by the thickness of GM and the hydrological variables including the magnitude of simulated event and antecedent moisture condition of GM. From the model development perspective, it is always desired to know the potential linkage between water quality and water quantity. Thus, the sensitivity/dependence of the EMCs of water quality parameters on the cumulative inflow, which is the sum of water volume applied to the laboratory cells from first experiment run to current experimental run, was investigated herein using linear regression approach for each GM type. The developed regression equations along with their coefficients of determination (R<sup>2</sup>) are displayed in Table 3. Sample scatter plots of the EMCs of three selected water quality parameters (including TN, NH<sub>4</sub><sup>+</sup> and Zn) and the cumulative inflow are demonstrated in Figure 2. As illustrated in the results, the EMCs of nutrients (except NH<sub>4</sub><sup>+</sup> of GM1 and GM3, and TP and PO<sub>4</sub><sup>3-</sup> of GM3) and Zn (except of GM3) decrease with the increase of the cumulative inflow. In contrast, the EMCs of Pb (except in GM1) increase with the increase of the cumulative inflow. No

consistent result on the dependence of the EMCs on the cumulative inflow was found for  $\text{NH}_4^+$  among the three GMs. Therefore, it can be concluded from the experimental study that the degree of leaching of nutrients (except  $\text{NH}_4^+$ ) in general decreases with time due to the increase of the cumulative inflow. The observed leaching behavior of nutrients and Zn is analog to the pollutant wash-off by stormwater surface runoff, which is related to the amount of pollutants available for wash-off on land surface. Whereas the leaching of Cu and Pb is either not significantly dependent on or significantly positively dependent on the cumulative flow. Thus the “wash-off” concept for the leaching of nutrients and Zn is not applicable for the leaching of Cu and Pb.

Table 2. Descriptive statistics of the EMCs of water quality parameters obtained in laboratory experiments and Kruskal Wallis test results followed by multiple comparison.

Water quality	GM type	Mean	Median	Standard deviation	Min	Max	Sample size	Comparison
$\text{NO}_3^-$ (mg/L)	GM1	72.50	42.50	51.55	30.00	170.00	8	GM1<GM2 GM3<GM2
	GM2	408.75	366.25	190.43	227.50	740.00	8	
	GM3	91.88	87.50	34.12	50.00	165.00	8	
$\text{NH}_4^+$ (mg/L)	GM1	11.63	11.50	4.34	7.00	21.00	8	GM2<GM1 GM2<GM3
	GM2	3.72	3.00	1.68	2.25	7.00	8	
	GM3	9.81	9.25	4.14	4.50	16.00	8	
TN (mg/L)	GM1	148.75	129.00	75.16	74.00	248.00	8	GM1<GM2 GM3<GM2
	GM2	682.69	615.00	273.36	325.00	1165.00	8	
	GM3	199.13	207.25	52.95	129.00	285.00	8	
$\text{PO}_4^{3-}$ (mg/L)	GM1	28.15	22.50	14.69	19.00	62.00	8	GM2<GM3
	GM2	15.69	13.50	4.19	12.25	23.50	8	
	GM3	66.97	64.58	12.05	52.60	90.00	8	
TP (mg/L)	GM1	28.31	23.83	9.01	22.00	46.00	8	GM1<GM3, GM2<GM3
	GM2	19.82	17.19	6.10	13.50	30.00	8	
	GM3	74.97	71.50	13.45	57.50	91.00	8	
Zn ( $\mu\text{g/L}$ )	GM1	131.25	90.00	89.67	50.00	250.00	8	GM1<GM2 GM3<GM2
	GM2	1655.00	1295.00	699.08	1010.00	2520.00	8	
	GM3	188.75	155.00	128.00	60.00	470.00	8	
Cu ( $\mu\text{g/L}$ )	GM1	1.63	1.00	2.07	n/a*	6.00	8	--
	GM2	1.88	1.50	1.64	n/a	4.00	8	
	GM3	1.43	1.00	1.27	n/a	3.00	7	
Pb ( $\mu\text{g/L}$ )	GM1	146.75	147.00	56.24	79.00	240.00	8	--
	GM2	259.86	291.00	141.56	103.00	404.00	7	
	GM3	228.43	200.00	144.88	63.00	422.00	7	

\* n/a: no analysis. The EMC could not be calculated because BDL (below detection limit) was reported in one or more than one samples in some simulated events.

Table 3. Regression equations (whose slopes are statistically significant) of the EMCs of water quality (y) on the cumulative inflow (x in the unit of  $\text{m}^3$ ) for each GM type

Water quality	GM type	Regression equation	$R^2$
$\text{NO}_3^-$ (mg/L)	GM1	$y = -3.77x + 162.93$	0.80
	GM2	$y = -14.53x + 757.10$	0.87

	GM3	$y = -2.01x + 139.99$	0.52
NH <sub>4</sub> <sup>+</sup> (mg/L)	GM1	--	--
	GM2	$y = -0.11x + 6.32$	0.63
	GM3	$y = 0.24x + 4.15$	0.49
TN (mg/L)	GM1	$y = -4.99x + 268.38$	0.66
	GM2	$y = -20.66x + 1,178.07$	0.86
	GM3	$y = -4.13x + 298.23$	0.91
PO <sub>4</sub> <sup>3-</sup> (mg/L)	GM1	$y = -0.87x + 49.02$	0.53
	GM2	$y = -0.21x + 20.78$	0.38
	GM3	--	--
TP (mg/L)	GM1	$y = -0.59x + 42.51$	0.65
	GM2	$y = -0.36x + 28.48$	0.52
	GM3	--	--
Zn (µg/L)	GM1	$y = -6.70x + 260.60$	0.82
	GM2	$y = -49.00x + 2,830.13$	0.74
	GM3	--	--
Cu (µg/L)	GM1	--	--
	GM2	--	--
	GM3	--	--
Pb (µg/L)	GM1	--	--
	GM2	$y = 9.59x + 26.41$	0.80
	GM3	$y = 9.98x - 29.13$	0.68

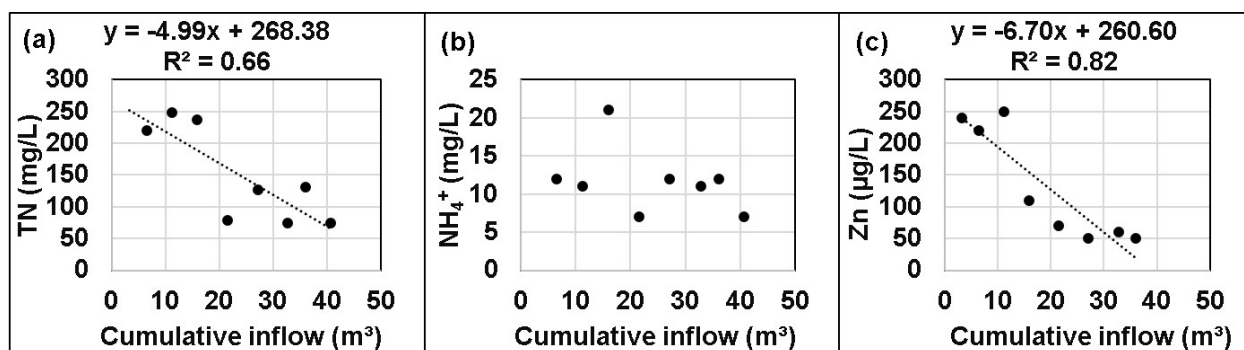


Figure 2. Scatter plots between the EMCs of TN, NH<sub>4</sub><sup>+</sup>, and Zn and the cumulative inflow for GM1. The linear regression lines (the dashed lines) whose slopes are statistically significant are included.

To investigate which GM is more sensitive to the cumulative inflow among the three GMs, the regression slopes which are statistically significant were compared for each investigated water quality parameter using the ANCOVA test followed by multiple comparison. The results are summarized in Table 4. No significant differences in the regression slopes were identified for PO<sub>4</sub><sup>3-</sup> and TP between GM1 and GM2; while these two pollutant constituents were found not to be statistically significantly dependent on the cumulative inflow in GM3 due to the absence of significant regression slopes (Table 3). As discussed previously, highest medians of the EMCs of PO<sub>4</sub><sup>3-</sup> and TP were observed from GM3. Thus, the contents of PO<sub>4</sub><sup>3-</sup> and TP in GM3 might be more than enough for wash-off so that their decreases did not show in the experiment study. In contrast, the EMCs of both TN and NO<sub>3</sub><sup>-</sup> of GM2 are statistically most sensitive to the cumulative inflow among the GMs; while the degree of sensitivity of their leaching to the cumulative inflow were detected to be similar between GM1 and GM3. The leaching of Zn from GM2 is more sensitive to the cumulative inflow

than that from GM1. The results demonstrate that these pollutant constituents leach at different rates among the three GMs, which might be ascribed to the chemical properties of the GMs. For instance, the leach rates of TN and Zn were statistically significantly higher in GM2, which has highest initial contents of TN and Zn. These results argue that the effect of chemical properties of GM on chemical leaching. Furthermore, compared to the leaching of TP, the leaching of TN appears to be faster from all three GMs, as the slopes of regression lines for TN are obviously higher than those for TP. It thus can be concluded that nitrogen is more mobile and thus can leach more easily from the GMs compared to phosphorus.

Table 4. Comparison of the statistically significant regression slopes among three or between two GMs in the laboratory experiments and between the laboratory cell of GM1 and the full-scale green roof (GR).

Comparison	Laboratory (GM1, GM2 & GM3)	Laboratory (GM1) and GR
NO <sub>3</sub> <sup>-</sup>	GM1<GM2, GM3<GM2, GM1=GM3	n/a*
NH <sub>4</sub> <sup>+</sup>	GM2<GM3	n/a
TN	GM1<GM2, GM3<GM2, GM1=GM3	n/a
PO <sub>4</sub> <sup>3-</sup>	GM1=GM2	GR <GM1
TP	GM1=GM2	GR <GM1
Zn	GM1<GM2	n/a
Pb	GM2=GM3	n/a

\* n/a: no analysis conducted due to the absence of statistically significant regression slopes detected in GR.

### 3.4 Chemical Leaching from the Full-Scale Green Roof

Several descriptive statistics of the EMCs of water quality parameters for GR and RR as well as the results from the comparison of the medians of the EMCs between GR and RR are presented in Table 5. When using the RR as the reference, the observations show that GR has acted as the source of nutrients since the commence of their operation in May of 2014 as all statistics of GR presented in Table 5 are obviously higher than those of RR. In contrast to nutrients, GR functions as the sink of all three metals as the statistics of metals of GR are lower or slightly lower than those of RR. Furthermore, the medians of the EMCs of all nutrients of GR were detected to be significantly higher than those of RR. The median of Zn EMCs of RR is significantly higher than that of GR; whereas no significant differences in the medians of the EMCs of Cu and Pb were detected between GR and RR. Therefore, it can be concluded that GR functions as the source of nutrients; while GR acts as the sink of Zn during the field study period.

Table 5. Descriptive statistics of the EMCs of water quality parameters obtained in field experiments for both GR and RR and detected significant differences in their medians between GR and RR using the Kruskal Wallis test. Sample size for the analysis for all water quality parameters is 8.

Water quality	Roof type	Mean	Median	Standard deviation	Min	Max	Comparison
NO <sub>3</sub> <sup>-</sup> (mg/L)	GR	2.26	1.75	1.84	0.81	6.33	GR>RR
	RR	0.82	0.75	0.50	0.19	1.93	
NH <sub>4</sub> <sup>+</sup> (mg/L)	GR	0.82	0.49	0.89	0.28	2.94	GR>RR
	RR	0.20	0.13	0.18	0.01	0.49	
TN (mg/L)	GR	4.20	3.62	3.32	1.40	11.21	GR>RR
	RR	1.81	1.63	1.12	0.71	3.91	
PO <sub>4</sub> <sup>3-</sup> (mg/L)	GR	1.32	1.21	0.94	0.43	3.36	GR>RR
	RR	0.27	0.26	0.11	0.12	0.45	

TP (mg/L)	GR	1.45	1.25	1.00	0.45	3.62	GR>RR
	RR	0.31	0.29	0.13	0.15	0.49	
Zn (µg/L)	GR	17.51	15.65	5.13	12.90	28.59	GR<RR
	RR	341.29	363.15	123.83	170.50	488.80	
Cu (µg/L)	GR	9.70	8.28	4.08	5.29	18.04	--
	RR	14.81	13.05	8.67	6.44	33.00	
Pb (µg/L)	GR	1.81	2.00	1.18	0.50	3.82	--
	RR	2.58	2.18	1.61	1.00	5.80	

As the GR and GM1 cell have same growing media and structure, the comparison between GM1 and GR was also conducted. It was found that the medians of the EMCs of all pollutant constituents (except Cu) of GM1 are significantly higher than those of GR. This result is under expectation as the observations from the laboratory experiments represent the initial stage of green roof; while the observations from the GR represents the GR during its age between 1 and 3 years. Therefore, the higher EMCs at the initial stage of green roof operation is speculated and leaching is reduced with time as illustrated in the experimental study, in which the EMCs of many constituents decrease with the increase of the cumulative inflow. Similar to the experimental study, it was observed that the EMCs of all nutrients and metals (except Cu) decrease with time, namely with the increase of the cumulative inflow. The decline (from 2003 to 2004) in nutrient leaching was also observed (TRCA, 2006; Harper *et al.*, 2015). The decline of chemical leaching of GR with time was also identified through comparing the EMCs of water quality parameters between 2015 and 2017, when field data are available. The medians of the EMCs of  $\text{NH}_4^+$ ,  $\text{PO}_4^{3-}$  and TP were found to be significantly higher in 2015 than those in 2017. On the other hand, opposite results were obtained for RR. Significant increases with time were observed in the EMCs of  $\text{PO}_4^{3-}$  and TP for RR. In addition, it was also observed that the EMCs of other water quality parameters appear to increase from 2015 to 2017 for RR. The observed increases of the EMCs of some pollutants for RR could be due to the change in atmospheric deposition which is the primary source of pollutants. These results demonstrate that the chemical leaching from GR in general decline with time, which is similar to the observations obtained from the experimental study. However, in the experimental study, Pb was shown to increase with the cumulative inflow from laboratory cells GM2 and GM3, which should be further investigated.

To investigate whether the progression of chemical leaching of GR can also be explained by the cumulative inflow, which was identified in the experimental study, the linear regression equations of the EMCs of water quality parameters on the cumulative inflow were also developed and are presented in Table 6. The cumulative inflow is the accumulation of precipitation (rainfall and snowfall) and irrigation water for GR and the accumulation of precipitation for RR since the start of their operation. As shown in Table 6, the significant regression slope was identified for TP,  $\text{PO}_4^{3-}$ , and Pb for GR; while it was identified for TP and  $\text{PO}_4^{3-}$  for RR. The results demonstrate that the leaching of these pollutant constituents can be largely explained by the cumulative inflow. However significant slopes were not detected in the regression equations for TN and  $\text{NO}_3^-$  from GR, which is different from the laboratory results (GM1). The removal efficiency of nitrogen of various LID (such as bioretention systems and green roofs) have been reported to be also associated with temperature (Buffam *et al.*, 2016; Van Seters *et al.*, 2009). The laboratory experiments were conducted at controlled and approximately constant temperature. Therefore, the role of temperature might be significant in the field. In addition, comparing the regression slopes, which indicate the leaching rates, between GM1 and GR (Table 4), both TP and  $\text{PO}_4^{3-}$  leach faster from GM1 than from GR, This, result implies that the decline of nutrient leaching could be slower in the late stage compared to the leaching at the beginning. Furthermore, the EMCs of TN and  $\text{NO}_3^-$  appear to be similar in magnitude in the events collected in 2017 (results not shown), which might explain the absence of the significant slopes for nitrogen for GR. All these results support that nitrogen leaches faster than phosphorous. Phosphate, which often is the major component of TP (approximately 90% in average in this study), can be retained in the ground by biological activities, absorption, and mineralization (Lehmann and Schroth, 2003).



Table 6. Regression equations (whose slopes are statistically significant) for the EMCs of water quality (y) on the cumulative inflow (x) for GR and RR, respectively. Note that the regression equations whose slopes are significant are presented here.

Water quality	Roof type	Regression equation	R <sup>2</sup>
PO <sub>4</sub> <sup>3-</sup>	GR	y = -0.004x + 3.73	0.53
	RR	y = 0.001x + 0.037	0.70
TP	GR	y = -0.005x + 3.99	0.52
	RR	y = 0.001x + 0.033	0.78
Pb	GR	y = -0.007x + 5.45	0.77
	RR	--	--

Although the observations from the GR could not be quantitatively explained by the observations from GM1, the qualitative results are similar in general between GR and GM1. In the field, the temporal behavior of nitrogen leaching might not be simply described by the cumulative inflow. However similar to pollutant wash-off by stormwater surface runoff, the chemical leaching especially nutrient leaching could be modeled based on the concept of pollutant wash-off combining with the other processes (e.g., biological and chemical processes).

#### 4 CONCLUSIONS

The laboratory experiments showed that all green roof cells (GM1, GM2, and GM3) are the source of pollution of both nutrients and metals in their early stage. The laboratory study results also demonstrated that the leaching of nutrients (except NH<sub>4</sub><sup>+</sup>) in general decrease with time as the cumulative inflow increases; however, their leaching responds to the cumulative inflow at different rates (the regression slopes) in different GMs. Furthermore, the degree of nutrient leaching is qualitatively associated with GM chemical properties. The leaching degree of metals appeared to not be related to their initial contents in GMs, and their leaching (except Zn) is either not significantly or significantly positively dependent on the cumulative inflow. The results supported that different physical processes govern the metal leaching and nutrient leaching, which could be modeled by pollutant “wash-off” process. From the field experiments, GR was identified to be the source of nutrients; whereas it acted as the sink of metals when using RR as the reference. The leaching of TP and PO<sub>4</sub><sup>3-</sup> from GR responded to the cumulative inflow similarly as observed in the laboratory experiments. However significant dependence between the leaching of TN and NO<sub>3</sub><sup>-</sup> and the cumulative inflow was absent, which might be explained by the important role of environmental variables (e.g., temperature). In addition, the comparisons between GR and GM1 and between GR in 2015 and GR in 2017 supported that the nutrient leaching is prominent in the very early stage of green roof establishment. Similar progression of the nutrient leaching was observed from both laboratory and field experiments. The results clearly showed that the progression of nutrient leaching would be primarily captured with the concept of wash-off combining with the other processes (e.g., biological and chemical processes).

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