Comparison of three methods for identification of building surface materials as sources of stormwater pollution

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Highlights

- Leaching experiments are effective in identifying sources of pollutants present in surface runoff
- Pollutant concentrations in surface runoff are best estimated by pilot-scale outdoor testing

Introduction

Surface materials of buildings and structures in urban areas were identified as potential sources of pollutants, which are released on contact with rainwater, and contribute to the pollution of urban stormwater (Müller et al., 2020). While the releases of metals from the metallic building envelopes (Malmqvist, 1983) and of biocides from e.g. renders and paints (Bollmann et al., 2014) have been well established in the literature, there is a scarcity of information on releases of other organic micropollutants. Several approaches to identifying surface materials as potential, or confirmed, sources of micropollutants were found in the literature. For instance, Björklund (2010) used a substance flow analysis (SFA) to identify which building surface materials were likely sources of phthalates and nonylphenols (NPs). Lamprea et al. (2018) performed leaching experiments with shredded pieces of automotive and construction materials and found several of them to be potential sources of bisphenol A and alkylphenols (APs). The sampling of residential runoff near Paris showed that building washoff was among the main contributors of APs to runoff (Bressy et al., 2011), but the specific materials releasing APs were not identified. In our study, three methods for identifying the surface materials contributing micropollutants to washoff were compared: (i) a material composition screening, (ii) laboratory leaching experiments and, (iii) outdoor pilot-scale testing of material sample panels. These methods served to identify potential sources of metals, APs and phthalates in urban runoff, and assess the strength of the identified sources. Furthermore, the pros and cons of individual methods and their applicability in future studies were discussed.

Methodology

Three methods were selected for studying the potential and actual releases of the targeted chemicals from a battery of tested materials: (i) The material composition screening was intended to identify potential pollutant sources by determining the chemicals present in the material composition; (ii) laboratory leaching experiments mimicked the releases of the targeted chemicals from materials tested on contact with lab water; and (iii) the outdoor pilot study was intended to test the materials studied for releases of the targeted chemicals under real-world conditions of actual rainfall/runoff events. In total, eight building surface materials were studied, and four of those were subject to all the three test methods. The materials studied and the tests applied are listed in Table 1.

Table 1. List and short description of the studied materials, and indication of which test methods they were subjected to.

Material	Label	Material composition screening	Laboratory leaching	Outdoor pilot study
Zinc sheet	ZN		X	X
Galvanised steel	GAL		X	X
Bitumen roofing felt	BIF	X	X	X
Bitumen roofing shingle	SHI	X	X	X
PVC roofing membrane(1)	PVCA	X		X
PVC roofing membrane(2)	PVCB	X	X	X
Coated, corrugated steel	CST	X	X	X
Copper sheet	CU		X	X

Material composition screening

The main objective of the material composition screening was to determine the actual presence (or absence) of phthalates and APs in the materials tested. Metal sheets were excluded, because it was highly unlikely that they would contain any organic micropollutants. For sample preparation, the coating of the coated, corrugated steel (CST) was scraped off, but the other materials, cut to an appropriate size, were treated as whole samples, because their surface layer could not be separated from underlying layers. One sample per material was prepared and sent for analyses to an accredited laboratory (ALS Scandinavia AB), where the samples were analysed by liquid chromatography–tandem mass spectrometry, LC-MS-MS, (for APs and their ethoxylates) and gas chromatography-mass spectrometry, GC-MS, (13 phthalates).

Laboratory leaching experiments

A synthetic rainwater with pH 4.4 was prepared according to the rainwater quality data from Swedish measurements and used as a leachant. The leachant was added to plastic beakers (for metal analyses) and glass beakers (organic analyses) to a volume corresponding to 32 mm, i.e., the rain depth of a 24 h rain event with one-year average recurrence interval in Sweden, after which the material samples, in duplicates, were immersed into the beakers. In order to limit the leaching to the sample surface that would be in contact with washoff on an actual building, the material edges and back face were covered with a metal-free lacquer (for metal analyses), or the samples were screwed back-to-back (for organic analyses). Control samples were made of plexiglass coated with the metal-free lacquer (metal analyses), and glass beakers without any material sample (organic analyses). After 24 hours of leaching under gentle agitation of 60 revolutions per minute, the leachates were sent to ALS Scandinavia AB for substance analyses, using inductively coupled plasma-sector field MS (ICP-SFMS) for metals, and GC-MS was used to analyse 10 phthalates, nonylphenols (NPs) and – ethoxylates (NPEOs). The laboratory leaching experiments were previously presented in Andersson Wikström et al. (2015).

Outdoor pilot study

Three replicates of every material were installed as 1 x 2 m mildly sloping panels outside at the campus area of Luleå University of Technology in northern Sweden. Luleå has subarctic climate (Köppen climate classification code Dfc). Stainless steel gutters were used to collect the runoff from each surface, and stainless steel was also represented on three panels; as a control material to monitor the contribution of pollution from the atmospheric deposition. All panels were placed in random order over the experimental site. For the purpose of this study, six rain events were sampled. The whole runoff volume from one panel was collected in one perfluoroalkoxy alkane (PFA) bag and treated as a single sample, giving event mean concentrations (EMCs). After each rain event, the samples were submitted to ALS Scandinavia AB for substance analyses: by ICP-SFMS (metals) and GC-MS (13 phthalates, APs and – ethoxylates). The pilot study was presented in detail in a previous publication (Müller et al., 2019).

Results and discussion

The results for selected substances are presented in Table 2. For brevity, the substances that were in all three sub-studies found to be below the reporting limits (RL), or not significantly different from the control samples (used in the laboratory and pilot studies), were excluded. In the material composition screening, PVCA and PVCB were identified as potential sources of the phthalates diisononyl phthalate (DINP), Diisodecyl phthalate (DIDP), and Di-n-octyl phthalate (DNOP), PVCA also contained Di-(2-ethylhexyl)phthalate (DEHP), and PVCB contained NPs. The PVC materials were both confirmed as sources of phthalates by the results of the pilot study, where especially the phthalates DINP and DIDP were detected in the runoff. In the laboratory leaching study, no phthalates were detected in concentrations above the RL. However, DINP and DIDP were not analysed for, and PVCA was not among the studied materials. Regarding NPs, PVCB was by all sub-studies identified as a source, and even though the NP concentrations were below the RL for BIF, SHI and PVCA in the material composition screening, NPs were still released from the materials in the other sub-studies. The concentrations of all the studied phthalates, NPs, NPEOs, octylphenols (OPs) and -ethoxhylates (OPEOs) in the surface layer of CST were below RL in the material composition screening. In the leaching study, ZN and GAL were identified as potential sources of Zn, and CU as a source of Cu; all of which were confirmed by the outdoor pilot study.

Table 2. Concentrations of selected substances in each sub-study (in material composition screening, the concentrations listed correspond to single samples; in laboratory leaching, the concentrations are the means of duplicate samples; and, in the pilot study, concentrations are the means of triplicate samples from six rain events. Where elevated RL were reported by the laboratory, instead of actual concentrations, such RL are presented in italics).

Material	Substance	Material composition screening (mg/kg)	Laboratory leaching (µg/L)	Outdoor pilot study (µg/L)
ZN	Zn	n/a	12,000	7770
GAL	Zn	n/a	9120	3530
BIF	DINP, DIDP	*<150*	n/a	n/a
	NPs	<10	2.0	0.99
SHI	Cu	n/a	43	24
	Ni	n/a	45	16
	NPs	<10	0.41	0.15
PVCA	DINP	190,000	n/a	365
	DIDP	*<30,000*	n/a	12
	DNOP	2700	n/a	1.7
	DEHP	770	n/a	1.0
	NPs	<10	n/a	0.23
PVCB	DINP	140,000	n/a	455
	DIDP	41,000	n/a	54
	DNOP	*<2000*	<0.6	2.0
	DEHP	*<100*	<1.3	<1.0
	NPs	34	8.9	26
CST	Zn	n/a	380	109
CU	Cu	n/a	1525	3090

* n/a = not applicable; the specific substance/material was not included in that sub-study.

The results of the leaching and the pilot studies were consistent with respect to identifying the substances released. Therefore, the leaching experiments, similar to those applied in our study, may be useful for identifying the sources of specific pollutant groups, with smaller resources required (i.e., in terms of the equipment, labour, safety, and costs). However, the leachate concentrations from such laboratory experiments did not agree well with the respective concentrations in runoff from the outdoor pilot panels, mimicking fairly well the actual building surfaces. Material leaching processes in the lab and the field differ in several aspects: material contact time, exposed surface area, temperature variations, dry periods (Lamprea et al., 2018), solar radiation, and the leachant liquid (i.e., actual, or synthetic rainwater). The material composition screening may serve as a good indicator of which pollutants could be present in the runoff, in a similar way as the SFA. However, our results indicate that such screenings may fail to detect some substances present in runoff and leachate samples, as was the case with NPs from BIF, SHI and PVCA.

Conclusions and future work

Laboratory leaching experiments are effective in identifying sources of substances present in surface runoff, but are not suitable for estimations of actual concentrations in such runoff. Material composition screenings requires the least resources, but may fail to identify minor sources of pollutants.

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