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Heavy metal contamination in surface runoff sediments of the urban area of Vilnius, Lithuania

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Abstract. Surface runoff from urbanized territories carries a wide range of pollutants. Sediments in untreated runoff from direct discharge stormwater systems significantly contribute to urban waterway pollution. In this study, heavy metal (Pb, Zn, Cu, Cr, Ba, As and Fe) contamination in surface runoff sediments of the urban area of the city of Vilnius was investigated. The surface runoff sediment samples were collected from seven dischargers with the highest volume rate of water flow and concentrations of suspended solids. The geospatial analysis of the distribution of heavy metals shows that there are several active pollution sources supplying the dischargers with contaminated sediments. Most of these areas are located in the central part of the city and in old town with intense traffic. Principal components analysis and *t*-test results clearly depicted the significantly different chemical compositions of winter and autumn surface sediment samples. The sampling approach and assessment of results provide a useful tool to examine the contamination that is generated in urban areas, distinguish pollution sources and give a better understanding of the importance of permeable surfaces and green areas.

Key words: dischargers, contamination, GIS, sediments, heavy metals.

INTRODUCTION

Surface runoff from urbanized territories carries a broad spectrum of pollutants. It is well known that urbanized areas are important pathways for the transfer of heavy metals (HM) into the environment (Buzier et al. 2011). Due to their dispersion, HM can be observed in different environments (Megateli et al. 2009; Drozdova et al. 2015). Polluted surface runoff may represent a short-term problem when dissolved pollutants enter a receiving system with pulses, and a long-term problem when toxicants accumulate in the sediments (Hatch & Burton 2000; Göbel et al. 2007).

Sediments in untreated runoff from direct discharge stormwater systems are one of the most important contributors to urban waterway pollution, and are considered to be a dominant stressor in urban aquatic ecosystems (Marshall et al. 2010). Excess suspended sediments can affect the aquatic ecosystem through depositional effects such as the reduction in the exchange capacity between benthic and water column zones, reduced food quality and smothering of biota, as well as through suspended effects such as respiratory damage, light attenuation and transport of other pollutants such as HM (Ryan 1991; Pekey 2006; Clapcott et al. 2011).

Urban runoff sediments are derived from several sources which include direct sources such as vehicle, tyre and brake wear, exhaust, oil spills, surface material degradation (weathering, renovation, demolition, paint) and soil erosion (Zanders 2005; Egodawatta et al. 2009; Wicke et al. 2012) and indirect sources such as atmospheric deposition (Murphy et al. 2014). Heavy metals are one of the most dangerous pollutants for aquatic environment. Heavy metals in surface runoff have a number of sources: construction sites, roofing, abrasion of automobile tyres, abrasion and corrosion of brake pads and other vehicle details, and combustion of fuel (Göbel et al. 2007; Camponelli et al. 2010).

Sediment-associated metals accumulate in the river during periods of low discharge; they are resuspended and transported downstream during flood events (Ciszewski 2001), especially during high flow periods. These high flow periods are caused by heavy rain or intense snow melt episodes, which can wash out large amounts of sediments into the river in a short period of time (Zabaleta et al. 2007). The ability of sediments to adsorb organic and inorganic contaminants makes sediment analysis a valuable tool to assess and monitor water quality and track contaminant transport in fluvial realms and mobilization in lake or marine environments (Hejabi & Basavarajappa 2013).

Contaminated sediments in an aquatic system can be the cause of secondary pollution via the geochemical circulation of pollutants as river sediments not only act as the main sink for HM. Due to the change in the aquatic environment it can pose serious ecological risks (Reza & Singh 2010; Martin et al. 2015). The level of contamination in the sediments shows the health of the ecosystem as a whole. Most of the cities encounter the problem with pollution. These cities are mostly located on rivers or in coastal areas. Thus these ecosystems get a quite high load of contaminants due to the urbanization and industrialization (Zhang et al. 2011; Staley et al. 2015).

Therefore there is a demand for more concern on sediment pollution, as many previous studies have confirmed an increased contamination level of sediments in rivers by HM in urban areas (Abuduwaili et al. 2015; Chen et al. 2016; Zhang et al. 2016). Many studies have revealed that most of the metals in aquatic systems are associated with the particulated phase. These pollutants are stored in fine-grained sediments and complexed to organic matter and oxides (Devesa-Rey et al. 2010; Owens & Xu 2011; Bartoli et al. 2012; Chen et al. 2016).

A total of 94 surface runoff discharges are located on the Neris River in the territory of the city of Vilnius. Most of these dischargers are not connected to treatment plants. The central part of Vilnius and the old town are the cultural heritage territories where treatment plants are not allowed to be established. Impervious surfaces such as roads, bridges, parking lots and buildings cause slow percolation of water into the ground. In this case, the water remains above the surface, accumulating and running off in large amounts and carrying large amounts of HM and other pollutants.

The studies conducted so far include only few sampling points of sediments directly in the river bed, while sediments in dischargers have not been investigated. It is the first time when sediments were taken for analysis directly from drainage pipes in Vilnius. The concentrations of HM in runoff sediments were quite high compared to those of Neris River sediments (Kruopienė 2007), but were similar to the HM concentrations in urban soils of Vilnius territory that are given in *Geochemical Atlas of Lithuania* (Kadūnas et al. 1999, pp. 121–125).

The main objective of our study was to analyse the load of contamination entering the river via sediments that form in urban areas and their seasonal fluctuations. We also aimed to evaluate the level of sediment contamination by HM and identify possible pollution sources.

MATERIAL AND METHODS

Study area and sample sites

Vilnius is the largest (capital) city in Lithuania with about 535 000 inhabitants. It is located in the southeastern part of Lithuania at the confluence of the Neris and Vilnia rivers (Fig. 1).

The catchment area for each discharger varies from 0.09 km^2 in the city centre to 10.72 km^2 in areas further from the centre (average catchment area is about 2.18 km^2). The GIS data of the catchment area were received from the municipal company ‘Vilnius plan’.

Land use categories (streets, buildings, forests, etc.) were analysed for each catchment using ArcMap 10.3.1 software and the geospatial data set (GDR10-LT). The highest percentage of impervious surface is characteristic of catchment areas that are located in the city centre (Table 1; Fig. 1). There it varies from 79% to 100% of the total catchment area, while in the periphery of the city it ranges from 44% to 71%.

Primary information about surface runoff dischargers was collected in the Lithuanian Environmental Protection Agency. Data included the results of dissolved oxygen, suspended solids, oil and its products, biochemical oxygen demand in 7 days (BOD7) and discharge. Seven surface runoff dischargers with the highest volume rate of water flow and concentrations of suspended solids were selected for the investigation.

Sample collection and analysis

Sediment samples were collected during the period of 29 September 2014 to 31 January 2015 from seven drainage pipes that carry storm and snowmelt runoff where sediments are accumulated. Six sampling expeditions were completed to collect three samples from each discharger immediately after storm events with high flow discharge (autumn period) and three samples during the thaw events with low flow discharge (winter period). A total of 42 samples were collected during the entire period of investigation. Sediment samples were placed in plastic bags and registered.

Samples were dried at 110 °C to constant mass. Then the particles of 125 μm size were separated and the concentrations of HM were analysed using a portative X-ray fluorescence spectrometer NITON

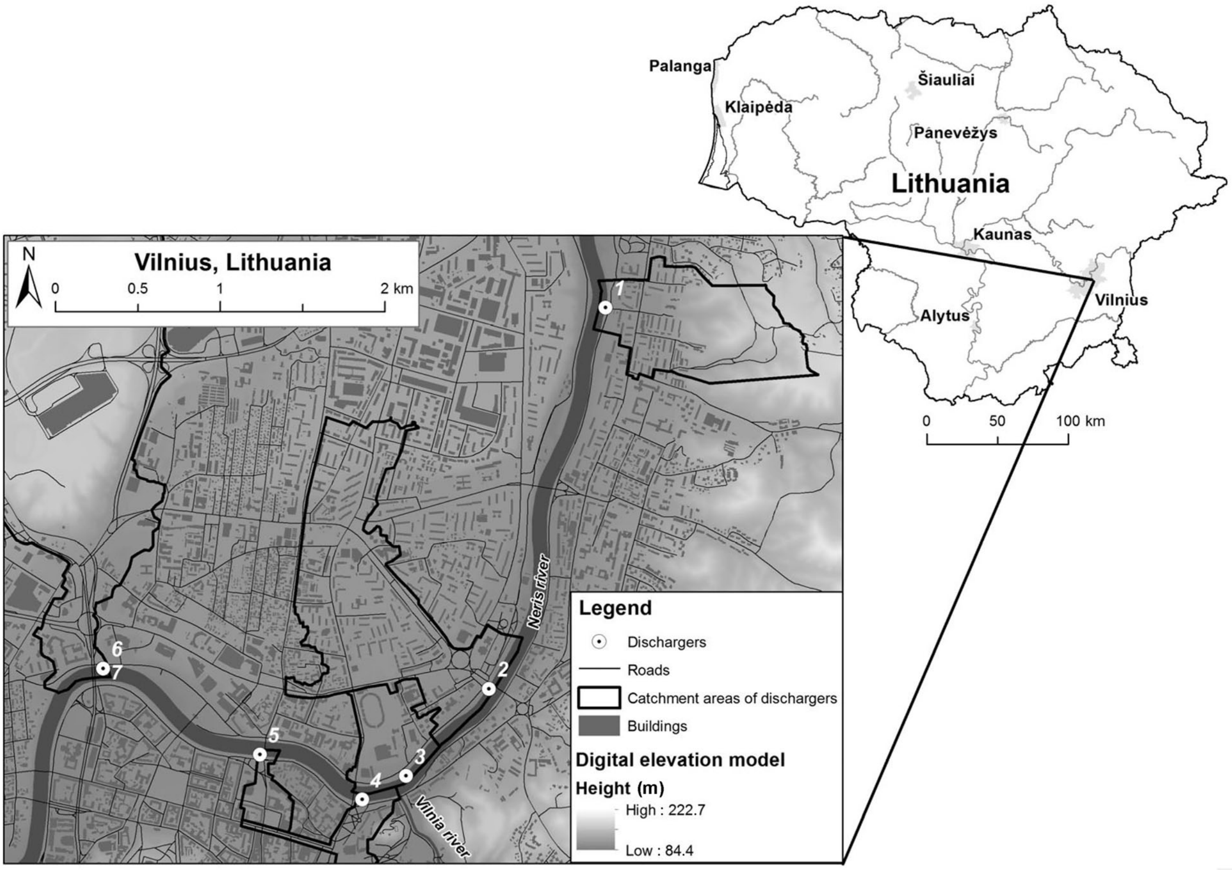

Fig. 1. Survey area and catchment areas for each discharger along the Neris River, Vilnius city centre.

Table 1. Distribution of permeable and impervious surface areas in each catchment area

No. of the catchment area	Area (km^2)	Permeable surface (%)	Impervious surface (%)
1	0.67	56.3	43.7
2	1.19	2.1	97.9
3	0.27	0.0	100.0
4	0.13	20.8	79.2
5	0.09	2.5	97.5
6, 7	10.72	28.9	71.1

XL2 Analyzer (2009). The total relative analytical error was within 5%. Fine sediment particles may be easily transported in suspension. Sediments up to 250 µm are not deposited and are carried through the catchment area and the discharger systems as suspended solids (Deletic et al. 2000; Djukic et al. 2016). The device was used only in stand at the laboratory to achieve the highest possible accuracy. The overall accuracy of chemical elements analysed is between 10% and 20%. Furthermore, this device was inter-calibrated with the atomic absorption spectrometer and the results were up to 20% of significant systematic difference.

Mathematical and statistical calculations were conducted using SPSS Statistics 17.0 and XLSTAT 2014 software. ArcMap 10.3.1 software was used for mapping and geospatial analysis.

RESULTS AND DISCUSSION

Heavy metals that were detected in 95% of the samples were chosen for further analysis. These include As, Cr, Cu, Pb, Zn, Ba and Fe. According to the concentrations of HM the samples were divided into two groups – autumn (high flow) and winter (low flow) period samples as the analysis showed quite significant differences between these two groups.

A high degree of variability and cumulative frequency distribution (see Fig. 2) are typical for surface runoff pollutants (Stone & Marsalek 1996; Brown & Peake 2006). These reflect the complex wash-off dynamics of

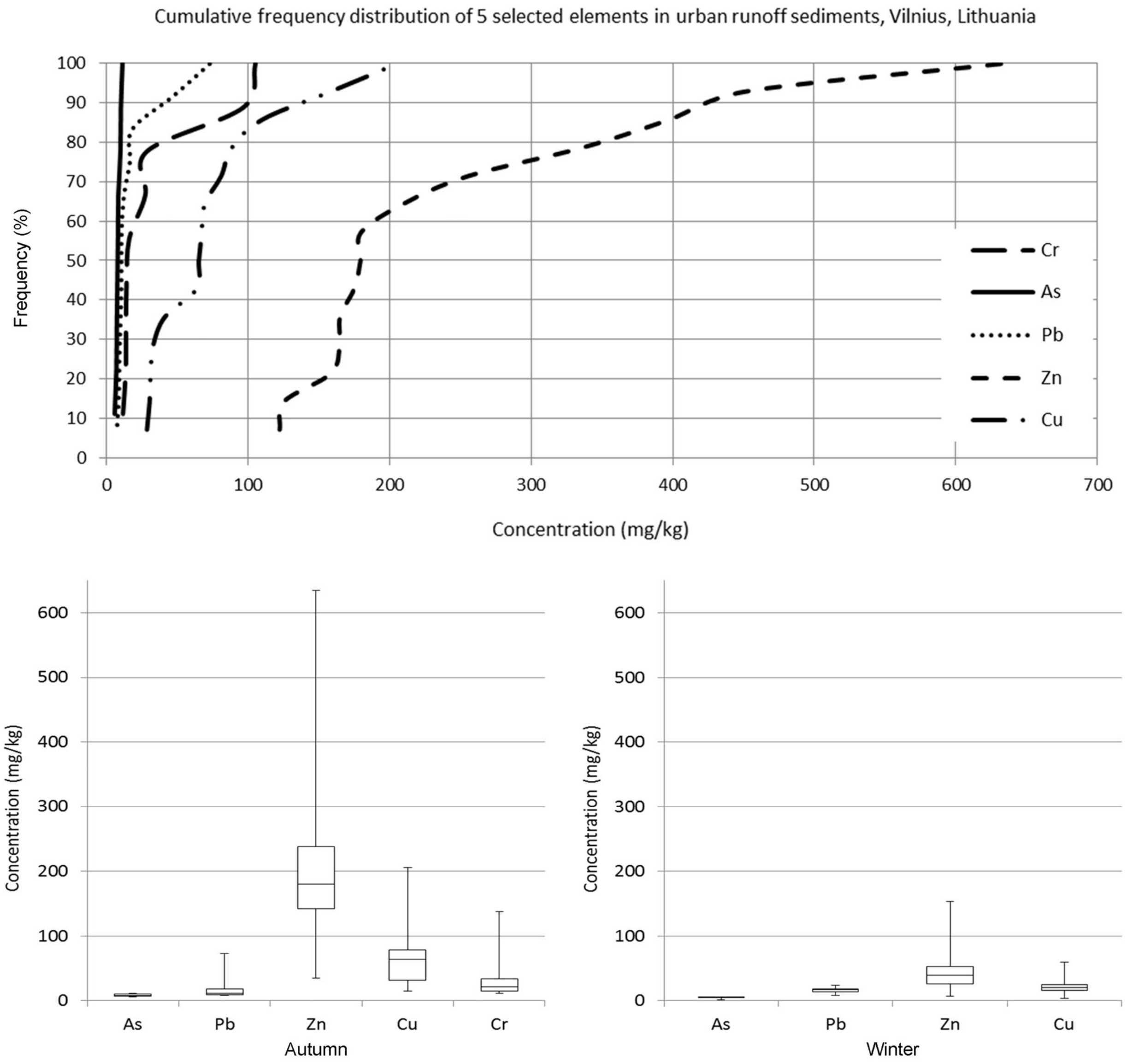

Fig. 2. Cumulative frequency distribution of Cr, As, Pb, Zn and Cu in urban runoff sediments of Vilnius ($N = 21$) and maximum, minimum and median values of their concentrations during autumn and winter periods.

the underlying surfaces and the variety of sources of the pollution within the urban catchment (Krein & Schorer 2000; Brown & Peake 2006).

Zn, Cu and Cr are characterized by the highest degree of variability in both autumn and winter periods. It is also clear that the autumn (high flow) period generates much higher concentrations of almost all HM than the winter (low flow) period. The concentration ranges of Zn, Cu, Pb and Cr were 35–635, 15–206, 8–73 and 12–138 mg/kg, respectively, in autumn samples. However, the concentration ranges of Zn, Cu and Pb were 20–153, 12–60 and 6–24 mg/kg, respectively, in winter samples. The concentrations of Cr were below the level of detection during the low flow period.

These results not only show the different wash-out dynamics of sediments and their chemical composition, but also the importance of permeable surfaces in urban territories. Soils are the major sink for HM released into the environment. The lack of green spaces in urban territories causes a more intense and faster access of HM to the river ecosystem. This is especially important for cities with large relative height differences as the geomorphology also contributes to a more intense transport of particles. The difference in height between the highest and lowest areas in Vilnius is 138.39 m, thus the results show the association between the transport of trace metals and sediments. This association is strong as the seasonal and annual transport of most trace metals can be estimated from sediment flux.

The highest concentrations of Zn and Cu exceed (2.1 and 2.7 times, respectively) the maximum allowable concentrations (MAC) that are stated for bottom sediments of surface water bodies (Table 2). The highest concentrations of Cr are close to the MAC, but do not exceed it. The Pb concentration does not exceed the MAC either. There is also a significant difference between autumn and winter samples. These contaminated sediments can be transported in large amounts during the high flow period and retained during the low flow period. In the low flow period sediments are washed with snowmelt water and thus filtrated to permeable surfaces. This process slows down the transportation of contaminated sediment particles to the river ecosystem.

High concentrations of Zn, Cu, Pb and Cr are found in areas such as old, central parts of Vilnius, small areas with intense traffic and the highest percentage of impervious surface (Table 1). These are dischargers 2, 3 and 4, and their catchments situated in the central part of the city. The highest Cr (76 mg/kg), Cu (159 mg/kg) and Pb (53 mg/kg) concentrations were detected in discharger 4. The concentration of Zn was slightly higher in discharger 2 (385 mg/kg) and reached 340 mg/kg in discharger 4. Only As concentrations are distributed inversely; the highest concentrations were detected in dischargers 6 and 7. These concentrations are quite low and thus can be attributed to natural sources or a very low anthropogenic impact for this element.

The lowest concentrations of HM of potentially anthropogenic origin were detected in dischargers 1, 6 and 7: Cr ranges from 14 to 22 mg/kg, Cu from 31 to 59 mg/kg, Pb from 8 to 13 mg/kg and Zn from 123 to183 mg/kg. Catchment areas (1, 4, 6, 7) are covered with a higher percentage of permeable surfaces (forests, grasslands and parks).

Table 2. The comparison of minimum, median and maximum winter and autumn Pb, Cr, Cu and Zn values (in mg/kg) with maximum allowable concentrations (MAC) in bottom sediments of surface water bodies (LME 2014). LOD – level of detection

		Pb	Cr	Cu	Zn
	MAC	<140	<140	<75	<300
Autumn	Minimum	8 ± 4	12 ± 8	15 ± 8	35 ± 7
	Median	11 ± 4	21 ± 8	64 ± 8	180 ± 7
	Maximum	73 ± 4	138 ± 8	206 ± 8	635 ± 7
Winter	Minimum	6 ± 4	<LOD	12 ± 8	20 ± 7
	Median	17 ± 4	<LOD	20 ± 8	40 ± 7
	Maximum	24 ± 4	<LOD	60 ± 8	153 ± 7

Our results show that it is very important to have enough area of green spaces and permeable surfaces in urban environment to mitigate the impact of pollution sources (e.g. intense traffic, oil spills, surface material degradation). The minimum percentage of permeable surfaces and distribution of green areas should be better implemented and clearly stated in spatial planning documents of urban areas.

Cumulative frequency distributions for Cr, As, Pb, Zn and Cu that are typical pollutants in urban environment are shown in Fig. 2. The cumulative distribution of heavy metal concentrations illustrates a relatively narrow range of concentrations. This means that high concentrations occur in a small percentage (up to 20%) of samples, thus indicating the presence of local active sources. However, the cumulative distribution of Zn and Cu concentrations shows a wider range of concentrations. High concentrations of these elements occur in a higher percentage (up to 30%) of samples which refer to more diffuse pollution sources (e.g. traffic, galvanized steel roofs). The average concentrations of metals in the sediment samples followed the order Zn > Cu > Pb > Cr > As.

Principal component analysis (PCA) provides information on the most significant parameters which explain the entire data set affording data reduction with a minimum loss of original information (Helena et al. 2000). It is an effective technique designed to transform the original variables into new, independent variables, called the principal components. In our study PCA was used to obtain an overview of the chemical data of the winter and autumn periods. Average concentrations of six elements (As, Ba, Pb, Zn, Cu and Fe) were calculated for each discharger separately for autumn and winter sediment samples. The first and second principal components described 88% of the total variation.

Figure 3 shows the loading plot which reveals the distinguished groups of samples with different chemical characteristics of autumn and winter periods. Most of the winter samples appear to correlate well as they occur in one group. Some of the autumn samples (dischargers A1 and A7) are quite near to the samples of the winter group. This is due to the similar concentrations of HM which appeared to be quite low. Most of the other autumn samples occur in one group as these dischargers transport sediments that are moderately polluted by HM.

The results of discharger A4 do not correlate with those of other dischargers because sediment samples that were taken from this discharger were the most contaminated by HM during the high flow period. Zn, Cu and Cr were the most important variables that

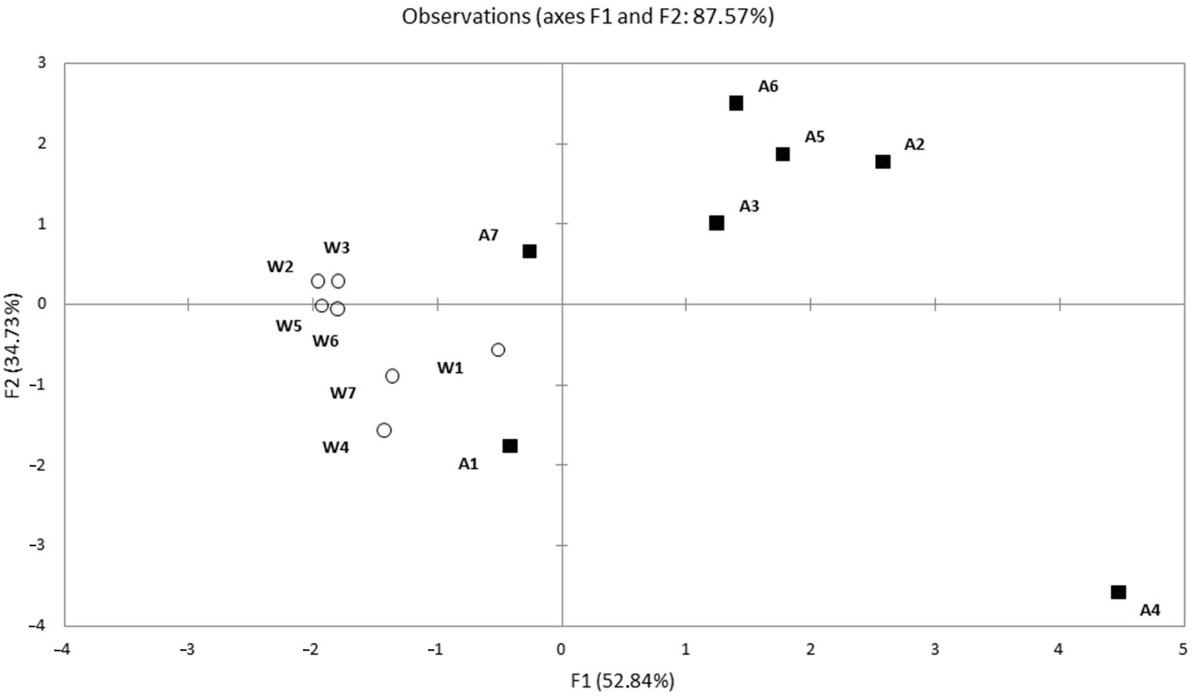

Fig. 3. Results of principal component analysis showing investigated concentrations of heavy metals during winter (W) and autumn (A) periods.

contribute to form discharger A4 as an individual between factors. The correlation matrix also showed a statistically significant (significance level $\alpha = 0.05$) correlation between Zn and Pb (0.768), Cu and Pb (0.947) and Cu and Zn (0.791). This clearly shows that these elements come from similar sources at the same period of time and contribute to the contamination of the river ecosystem. However, As and Ba inversely correlate or do not correlate with Pb, Cu and Zn.

To test the hypothesis that winter samples and autumn samples were associated with statistically significantly different HM concentrations, an independent samples *t*-test was performed. All the values (42 values of HM concentrations) of autumn and winter samples were included into *t*-test statistics.

Statistically significant values were obtained for As ($t = –1.866$ with $p = 0.073$), Pb ($t = –2.789$ with $p = 0.014$), Zn ($t = –4.888$ with $p = 0.000045$), Cu ($t = – 3.929$ with $p = 0.001$), Fe ($t = –6.626$ with $p = 0.000007$), Mn ($t = –3.704$ with $p = 0.002$). These results also confirm the results of PCA – the autumn (high flow) period is very important for river environment. This period provides the highest load of contaminants to the river ecosystem. In this respect dischargers should be constructed to minimize the access of polluted sediments, which should be better treated of removed. Urban planning should also properly contribute to reducing this problem (e.g. building green areas, more permeable surfaces).

Land use is the most important factor causing the elevated concentrations of HM in topsoils as well as in sediments of surface runoff in urban areas. The highest concentrations of Pb occur close to major road junctions on roads with high traffic. The levels of Pb are also high in the areas with the oldest housing. The highest concentrations of HM, Zn in particular, are generally found in city areas of both historical and contemporary industrial activity (Kelly et al. 1996; Bullock & Gregory 2009).

CONCLUSIONS

The approach outlined in this paper presents a possibility of revealing sources of surface runoff pollution by contaminants generated in urban environment and provides a basis for effective management of pollutants that enter the river ecosystem from urban territories. Detailed study of deposited sediments from urban surface runoff dischargers showed that there are sources within the impervious urban area supplying surface runoff with particle-bound pollutants such as HM.

Geospatial analysis clearly revealed the most important pollution sources in the city of Vilnius. High concentrations of Zn, Cu, Pb and Cr were mainly derived from anthropogenic sources and were found in areas such as old, central parts of Vilnius, small areas with intense traffic and the highest percentage of impervious surface. The highest concentrations of Zn and Cu exceeded the maximum allowable concentrations for bottom sediments of surface water bodies in Lithuania 2.1 and 2.7 times, respectively.

Principal component analysis and *t*-test results also exhibited statistically significant differences of chemical composition between winter and autumn surface runoff sediment. The high flow period (autumn) is crucial for the transportation of contaminants as HM come from similar sources at the same period of time and contribute to the contamination of the river ecosystem.

The process of water retention during the winter period is important for slowing down the transfer of contaminants. These processes are especially relevant for cities that are situated in complex geomorphological conditions with a low proportion of areas of permeable surfaces (green areas).

The approach explained in this paper enables establishing surface runoff pollution sources and contaminants that are generated in urban environment. It provides a basis for urban planning to better implement measures for the detention and purification of pollutants that get into the river ecosystem.

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Raskmetallid Vilniuse linnakeskkonna pindmise äravoolu setendites

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Linnakeskkonna pindmise äravooluna liikuv vihma- ja sulamisvesi ning sellega edasikanduvad setted on tihti saastunud erinevate keskkonnaohtlike komponentidega. Käesolevas uurimuses on selgitatud raskmetallide (Pb, Zn, Cu, Cr, Ba, As, Fe) esinemist ja jaotumist Vilniuse linnakeskkonna pindmise äravoolu vees ning sellega edasikanduvates setendites. Kõige enam on raskmetallidega saastunud sademete äravoolusetted suure liikluskoormusega kesklinnas, sealhulgas ka Vilniuse vanalinna piirkonnas, kus Zn ja Pb kontsentratsioonid ületavad mitmekordselt veekogude põhjasetetele kehtestatud piirväärtusi. Põhikomponentide analüüs näitab statistiliselt olulist erinevust suve-sügise ja talve pindmise äravooluga suspendeeritud hõljumi raskmetallide sisaldustes ning suuremad kontsentratsioonid on seotud suve-sügisperioodi tugevate vihmavalingute-tormidega. Uuringu tulemused annavad aluse sademete äravoolusüsteemide planeerimiseks ja linnakeskkonnast mobiliseeritud saastumise puhastamiseks enne, kui see jõuab looduskeskkonda.