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EFFICIENCY OF HEAVY METALS REMOVAL DURING ROOF RUNOFF INFILTRATION THROUGH VEGETATED SOIL

Stormwater discharged from urban areas can contain significant amounts of heavy metals, especially zinc and copper. Roof runoff is very often directed into the soil and may pose a risk for groundwater quality. Lysimeter research has shown that zinc and copper contained in stormwater may be effectively removed by passing through vegetated soil. The average removal efficiency was greater than 90% and did not depend on the metal concentration in the influent, and the concentrations in the effluent met the limits for drinking water. Infiltration may be a reliable treatment method for roof runoff and may make stormwater safe for groundwater recharge.

1. INTRODUCTION

Urban development results in land use modification through the increasing of impervious surfaces such as streets, sidewalks, parking lots, and roofs of buildings. During wet weather, stormwater is collected from these surfaces with washed-off pollutants and transported by combined sewer systems to wastewater treatment plants, or by drainage systems directly to receiving waters. These solutions may cause many problems in cities and urban ecosystems such as overloading drainage systems and small urban rivers, flooding, and the deterioration of water quality.

Stormwater runoff usually contains significant amounts of suspended solids. Other contaminants may exist as dissolved species, colloids, or particulates [1–7]. In Poland, according to the Regulation of the Minister of Environment (Journal of Laws No. 20060169), the concentration of total suspended solids in stormwater discharged into receiving waters and the ground should not exceed 100 mg/dm^3. Aside from this parameter, the regulation limits only the concentration of petroleum hydrocarbons – at the level of 15 mg/dm^3. Stormwater runoff may also contain significant quantities of

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heavy metals, mainly zinc, copper, lead, and cadmium [2–7]. The major anthropogenic sources of heavy metals are mining, smelting, electroplating, use of pesticides and fertilizers, industrial discharge, and atmospheric deposition [8] but in urbanized areas the most significant sources are vehicles and buildings. Numerous studies indicate that stormwater collected from roofs with metal elements (roofing materials: galvanized steel, copper sheets, and zinc gutters) often contains very high concentrations of zinc and copper due to the corrosion of metallic parts [6, 7, 9]. According to Zorbrist et al. [7], the corrosion rate of cooper in drains is about 5 $g\cdot m^{-2}\cdot y^{-1}$. Gromaire et al. [9] estimate that in Paris, zinc roofs produce from 33 to 60 t of zinc per year. Heavy metals such as zinc, cadmium, chromium(VI), copper, nickel, lead, and platinum are among the priority pollutants in urban stormwater that may impact the environment, particularly receiving waters [4]. The concentration of stormwater pollutants including heavy metals depends on several factors such as the location of the precipitation, rainwater pollution, catchment exposure, antecedent dry period, the duration and intensity of precipitation.

Recently, stormwater management has been increasingly implemented on the site of precipitation, mainly by directing it into the ground. Runoff is managed by structural best management practices (BMPs) – also called low impact development (LID) or sustainable urban drainage systems (SUDS) – such as infiltration trenches, biofiltration systems, rain gardens, and sand filters. Infiltration of stormwater runoff may be used as an alternative or supplement to conventional sewer systems in urbanized areas. This method of stormwater management complies with the principles of sustainable development, provided that water does not contain pollutants that may cause groundwater contamination. It is therefore necessary to identify potential sources of pollution and to use effective methods of stormwater treatment to remove contaminants potentially harmful to the environment.

Use of infiltration facilities with or without vegetation reduces the volume of stormwater runoff and removes contaminants, including heavy metals. The treatment is based on three basic mechanisms: filtration, sorption, and chemical and biological transformation and can be effectively optimized by the choice of infiltration media, control of system hydraulics, and control of redox conditions within the filtration system [5]. The metal mobility in the soil and bioavailability depend on the granulometric composition, organic matter forms, sorption capacity, cation exchange capacity, oxidation-reduction potential, and the activity of microorganisms [10]. pH of soil is one of the most important factors affecting the solubility or retention of metals [10, 11]. The most mobile metals are Cd, Zn, and Mo, while the least mobile ones are: Cd, Ni, and Pb [10]. Removal efficiency of heavy metals in infiltration facilities may be enhanced further by properly selected plants used in phytoremediation. These plants are characterized by the ability to grow in a contaminated environment and a high degree of accumulation or biodegradation of pollutants [8, 11, 12]. The bioavailability of metals is an important parameter conditioning the effectiveness of phytoremediation. Cd, Ni, Zn, As, Se, and Cu are easily absorbed metals, Co, Mn, and Fe are averagely absorbed, and Pb, Cr, and

U are poorly absorbed [8]. Research conducted by Sun and Davis [13] showed that 88–97% of metals were captured in soil media, 2.0–11.6% not captured by biofiltration and 0.5–3.3% accumulated in plants. The percent removal trend of the metals was as follows: Cd > Pb > Zn > Cu, and the accumulation in plants Zn > Cu > Pb > Cd, which happened to be the same regarding precipitation. In addition to the parameters of the filter layer and the type of plants, the conditions under which they operate influence the effectiveness of the facilities. Extended dry periods in excess of 3 weeks worsen significantly the biofilter metal removal performance [14]. Temperature and salt, and their interaction had an effect on outflow concentrations of metals. Temperature does not play a major role in biofilter performance, and no significant differences were observed between 4.6 and 17.1 °C but salt does have a negative impact on heavy metals removal, especially dissolved Cu and Pb [15]. Laboratory research and assessment of the BMP demonstrate a high efficiency of the removal of heavy metals during passage through the soil, vegetated or not but it does not mean that in all conditions there is no risk of groundwater contamination. Depending on the metal and its phase, the hydraulic parameters and the type of filling, metals can be retained in the upper layer of the filter, or also (sometimes later) can penetrate into the lower layers and the groundwater [2, 3, 5, 16–18].

In Łódź, Poland, in areas outside the central districts, and especially in single-family home residential areas where the roofs are usually covered with galvanized metal sheet or contain other metal elements, roof runoff is often discharged onto the lawns and then into the soil. Stormwater in the city, when discharged from the roofs, is characterized by high concentrations of metals, especially zinc [19, 20]. This paper presents the results of research on the efficiency of heavy metals removal in the vegetated soil which may allow an estimation of groundwater contamination potential from roof runoff infiltration practices.

2. EXPERIMENTAL

The research was conducted in the years 2012 and 2013 using 6 lysimeters (40×40 cm and 60 cm high) filled with natural soil to a depth of 40 cm. The lysimeters were planted with two species of plants: three with garden grass and three with *Deschampsia caespitosa*. The lysimeters were placed outdoors in positions of good sunlight, and additionally, after the precipitation, were watered with rainwater collected from roofs with various rooftop composition.

Rainwater collected from roofs covered with copper sheet was supplied to one lysimeter planted with grass and one planted with *Deschampsia caespitosa*. Likewise, rainwater collected from roofs covered with galvanized sheet was similarly supplied also to one lysimeter planted with grass and one planted with *Deschampsia caespitosa.* Two control lysimeters were watered with rainwater which had not been in contact with metallic materials. Rainwater was collected from various phenomena of precipitation (of different intensity and duration, and after various periods of dry weather) and from roofs of various age and state. During research, each lysimeter was watered periodically with ca. 10 l of rainwater, which corresponds to the 1 m^2 surface watered with rainwater collected from a 6.25 m^2 roof during a rainfall of 10 mm, thus hydraulic conditions were similar to those used in infiltration systems. Samples of influent and effluents were collected and, after pH measurements, digested with 65% HNO_3 in microwave digestion (MARSXpress by CEM Company). In each sample the concentration of four metals (zinc, copper, lead, and cadmium) was determined by the atomic absorption spectrophotometry (AAS, SOLAAR M5, Thermo Electron Corporation) in accordance with the Polish and European Standards. The flame module was used for Zn and Cu, and the graphite furnace module for Pb, Cd, and Cu (for lower concentrations). Metal removal efficiency (%) was calculated as follows:

$$\text{Removal efficiency} = \left(1 - \frac{\text{effluent concentration}}{\text{runoff concentration}}\right) \times 100\%$$

Atmospheric deposition was not taken into account. Metal concentration was also determined in harvested plants (in leaves and seeds) by AAS, after digestion with HNO_3. The scheme of the research is presented in Figs. 1 and 2.

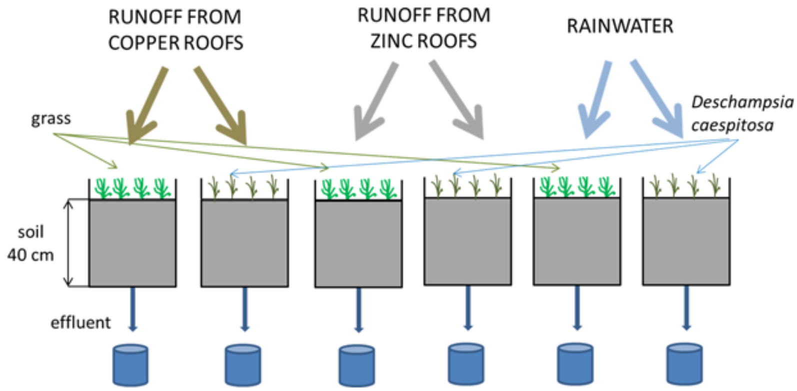

Fig. 1. Lysimeters combination characteristics

Fig. 2. The position of the lysimeters (left) and plants in the lysimeters (right)

3. RESULTS AND DISCUSSION

The concentrations of heavy metals in the roof runoff used in the research are given in Table 1.

Table 1

Concentration of heavy metals in the roof runoff (data from research and references)

Roofing material		Concentration [μg/dm³]			
		Zn	Cu	Pb	Cd
Year 2012					
Cooper sheet	range	20–350	1500–4600	4.1–13.4	<0.1–0.1
	mean	140	2960	8.2	0.1
Galvanized sheet	range	660–17200	30–1040	5.8–14.7	0.1–1.5
	mean	6430	480	9.9	0.5
Year 2013					
Cooper sheet	range	248–708	1083–6013	3.4–25.6	<0.1–0.7
	mean	444	3191	10.9	0.3
Galvanized sheet	range	1210–3923	5–26	0.71–15.5	0.1–0.4
	mean	1861	12	8.1	0.2
References					
Zawilski et al. [21]	range	41–4180	23–320	<2–130	<0.2–35.7
Langeveld et al. [2]		30–11700	6–500	2–404	–
Grebel et al. [5]		<13000	<1800	<525	–
Zgheib et al. [22]		130–520	30–220	<10–129	–
Chang et al. roofs [6]		39–212330	1–5410	25–700	–

Tables 2 and 3 present their concentrations in effluents from lysimeters planted with grass and *Deschampsia caespitosa.* Rainwater used in the research was collected from several roofs, so the concentrations and chemical forms of heavy metals were normal for these types of infiltration practices. As shown by numerous studies, the phases and compounds in which heavy metals are present in water have a significant impact on their mobility in the ground and assimilation by plants [3, 17, 21].

The data indicates that the concentration of heavy metals was varied, which was also confirmed by earlier studies [19, 20]. Usually high concentrations of heavy metals in roof runoff were observed, especially of zinc and copper (Table 1). They were sometimes several times higher compared to stormwater from urban drainage systems. Limits of metal concentrations in the stormwater discharged into the soil are not defined in Polish legal regulations so the results of the research can only be compared to the limits for drinking water in accordance with the Regulation of the Minister of Health (Journal of Laws 20100466), the requirements for industrial wastewater discharged into receiving waters and soil in accordance with the Regulation of the Minister of Environment (Journal of

Laws 20090169), as well as with the criteria of evaluation of groundwater in accordance with the Regulation of the Minister of Environment (Journal of Laws 20080896) (Table 4).

Table 2

Concentration of heavy metals in the effluent from lysimeters planted with grass

Lysimeters inflow		Concentration [μg/dm^3]			
		Zn	Cu	Pb	Cd
Year 2012					
Cu	range	30–300	60–680	3.61–27.5	<0.1–1.4
	mean	170	360	20.5	<0.7
Zn	range	110–2280	<0.5–40	9–43.8	0.2–1.9
	mean	600	<12	19.4	0.7
Rainwater	range	50–310	<0.5–20	7.9–15.1	0.1–0.4
	mean	140	<10	11.5	0.2
Year 2013					
Cu	range	44–157	27–39	6.7–28.1	0.1–0.6
	mean	97	31	14.4	0.4
Zn	range	36–295	7–46	3 –15.8	<0.1–0.4
	mean	108.10	18	9.0	<0.2

Cu – runoff from roof covered with cooper sheet.
Zn – runoff from roof covered with galvanized sheet

Table 3

Concentration of heavy metals in the effluent
from lysimeters planted with *Deschampsia caespitosa*

Lysimeters inflow		Concentration [μg/dm^3]			
		Zn	Cu	Pb	Cd
Year 2012					
Cu	range	70–210	50–850	2.5–28.9	0.1–0.8
	mean	140	270	16.46	0.43
Zn	range	80–440	<0.5–270	2.8–18.3	0.1–0.7
	mean	180	<140	11.51	0.5
Rainwater	range	70–120	<0.5–110	4.3–15.3	0.1–0.4
	mean	100	50	9.8	0.2
Year 2013					
Cu	range	23–80	14–101	5.5–20.6	0.1–0.4
	mean	56	35	11.7	0.2
Zn	range	60–131	11–15	3.1–27.5	0.1–0.4
	mean	98	13	12.7	0.2

Cu – runoff from roof covered with cooper sheet.
Zn – runoff from roof covered with galvanized sheet.

The concentration of zinc in drinking water is not defined in Polish regulations. According to the US EPA, the maximum concentrations of zinc for drinking water is 5000 μg/dm^3. The same level is recommended by WHO.

Table 4

Heavy metal concentration (median for years 2012–2013)
in view of water and wastewater criteria in Poland

Water/wastewater	Concentration [μg/dm^3]			
	Zn	Cu	Pb	Cd
Runoff from roof covered with cooper sheet	299	2959	7.76	0.1
Effluent from lysimeters with grass	73.9	80.3	12.8	0.2
Effluent from lysimeters with *Deschampsia caespitosa*	138.4	44.3	14.8	0.3
Runoff from roof covered with galvanized sheet	1477	23.1	9.5	0.2
Effluent from lysimeters with grass	118.9	14.6	12.4	0.2
Efluent from lysimeters with *Deschampsia caespitosa*	114	28.0	12.6	0.2
Drinking water	–	2000	25	5
Industrial wastewater discharged into receiving waters and soil	2000	500	500	400
Groundwater of good chemical quality	1000	200	100	5

The comparison of the heavy metal concentrations in roof runoff to the requirements for drinking water shows that the concentrations of copper in the runoff from roofs covered with copper sheet may be much higher than the permitted level. However, the concentrations of lead and cadmium are not exceeded, except the case of slightly exceeding the permissible concentration of lead. The concentration of zinc (not limited in drinking water) in the runoff from roofs covered with galvanized sheet, and copper in the runoff from roofs covered with copper sheet, may be much higher than the limits for industrial wastewater discharged into receiving waters and soil, and for groundwater of good chemical quality. The runoff from roofs containing metal components can thus pose a risk to groundwater quality and endanger drinking water resources, as was also emphasized by other authors [6, 7, 17].

Passing rainwater collected from roofs covered with copper or galvanized sheet through vegetated soil caused an effective reduction of the concentration of both copper and zinc. In the case of the lysimeters planted with grass, the average concentration of zinc and copper was lower than those required for industrial wastewater discharged into water and soil, and for groundwater of good chemical quality. However, in some phenomena, the limits were exceeded, as was the case with lysimeters planted with the *Deschampsia caespitosa* but here only the limit of copper concentration was exceeded. In the second year of research, there were no exceedances of permitted concentrations of zinc and copper in any sample. Concentration of the metals in the effluent from lysimeters was lower than those required for drinking water, aside from one case of exceeding the limit for lead. In the first year of the research, metal concentrations in the effluent from lysimeters were higher than those in the effluent from control lysimeters watered with rainwater, but in the second year of research concentrations of both metals in the effluent from lysimeters watered with roof runoff were comparable with effluent from lysimeters watered with rainwater. There were no significant differences between lysimeters planted with grass and *Deschampsia caespitos* which belongs to the species used in the process of phytostabilization [23, 24]. Plants used in phytostabilization can immobilize heavy metals in soils especially effectively, through sorption by roots, precipitation, complexation or metal valence reduction in the rhizosphere [8, 11].

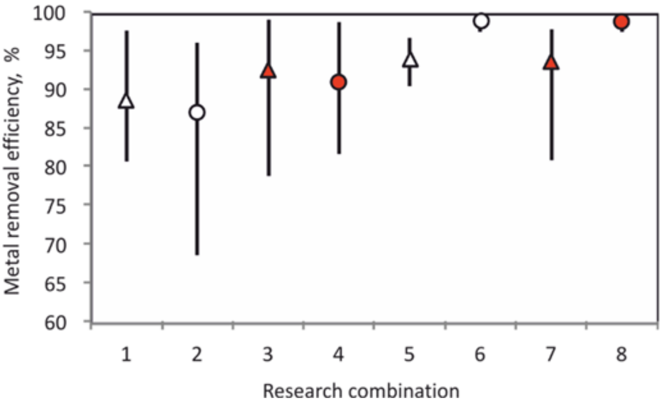

Fig. 3. Removal efficiency of zinc (triangles) and cooper (rings) in lysimeters planted with grass (empty) and *Deschampsia caespitosa* (solid). Mean and range (min–max) in 2012 (1–4) and 2013 (5–8)

In this study, the average removal efficiency of copper in the runoff from roofs covered with copper sheet was 93% in lysimeters planted with grass and 95% in lysimeters planted with the *Deschampsia caespitosa*, while zinc removal efficiency of the runoff from roofs covered with galvanized sheet was 91% in lysimeters planted with grass and 93% in lysimeters planted with the *Deschampsia caespitosa* (Fig. 3). As indicated in numerous studies, metal removal efficiency not only depends on the efficiency of the accumulation in plants, but also on the effectiveness of the metal retention in the soil, which is the highest in the upper layers [13, 14, 17]. The conducted research shows that the soil layer in the lysimeters 40 cm thick was sufficiently effective. In each of the lysimeters the differences in the effectiveness of the metal removal were observed in individual assays. It was probably due to a number of factors, including the degree of soil moisture associated with the length of the period of dry weather before testing, and pH, which was varied between 5.2 and 7.0. According to Ghosh and Singh [11], pH seems to have the greatest effect of any single factor on the solubility or retention of metals in soils, which was also confirmed by other studies [10, 21]. Usually the uptake of metals by plants decreases upon pH increasing to about 6.5–7.5. However, the effect of pH on metal mobility is highly variable, depending on the content and type of organic matter.

In the second year of studies, the metal removal efficiency was slightly higher and more stable, and only in one case was it lower than 90%. The increase in efficiency could certainly be affected by the growth of plants, especially their roots, and the stabilization of the substrate soil. The concentration of metals in the influent did not significantly affect the effectiveness of metal removal (Fig. 4).

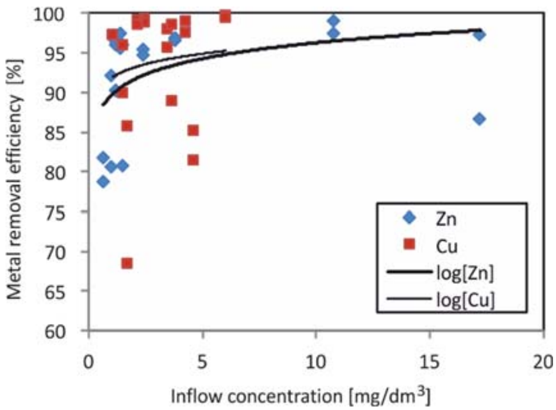

Fig. 4. Removal efficiency of zinc in the runoff from roofs covered with galvanized sheets and copper in the runoff from roofs covered with the copper sheets depending on their initial concentration in the inflow to the lysimeters

Table 5

Concentration of heavy metals in plants

Roofing material	Plant species	Concentraton [mg/kg DM]			
		Zn	Cu	Zn	Cu
		Seeds		Leaves	
Cooper sheet	grass	55.0	7.0	48.9	6.6
	Deschampsia caespitosa	53.2	8.6	33.3	5.8
Galvanized sheet	grass	45.7	4.8	40.1	6.8
	Deschampsia caespitosa	57.8	7.5	26.8	4.6

The concentration of heavy metals in plants (Table 5) was lower than those observed in other studies [8, 25]. A higher accumulation of zinc than copper in both plants was measured, as was the case of other research [13]. The ratio of zinc to copper in plants was similar in all the lysimeters, also in lysimeters watered with rainwater from the roof covered with copper sheets, wherein the copper concentration was several times higher than zinc. The concentration of the metals was higher in seeds than in leaves of both species of plants. According to Wołejko et al. [21], the metal concentrations in above--ground parts of grass is correlated with the concentrations in the surface layer of the soil. Metal concentrations in plants from lysimeters watered with runoff from roofs were not significantly higher than in plants from lysimeters watered with rainwater. It suggests that metals were retained in soil or in roots, as it happens during phytostabilization. Determination of metal accumulation in the soil and in the roots and shoots of the plants requires further study. This is important because of limited capacity of the system to store the metals. The method of heavy metals removal from the facilities depends on the place where metals are mainly stored [25].

4. CONCLUSIONS

Rain runoff from roofs with metal elements (roofing materials, gutters) may contain significant quantities of heavy metals, often many times greater than in the case of other impervious surfaces. Concentration of these metals is highly variable, depending on the state of the roofing and precipitation parameters.

Direct discharge of roof runoff into the soil may endanger the groundwater quality because zinc and copper concentrations may exceed the limits for drinking water and groundwater of good chemical quality.

Infiltration through vegetated soil allows a very effective removal of zinc and copper from stormwater, regardless of the initial concentration. There were no significant differences between grass and *Deschampsia caespitosa*, thus it can be assumed that the typical solution for the management of roof runoff – discharging onto garden lawns – does not pose a threat to groundwater quality.

Infiltration, particularly through vegetated soil, has the potential to provide reliable treatment of roof runoff and may result in stormwater that is safe for groundwater recharge, but where there are large surface areas of roofs covered with galvanized or copper sheets, and a high level of groundwater, it should be monitored to protect the water resources.

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