Analysis of concentration trends and origins of heavy metal loads in stormwater runoff in selected cities: A review

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Abstract. The currently heavy metal pollution in rainwater is a huge problem, especially in industrial areas. Sediment from devices for stormwater runoff treatment, especially those located in city centers, industrial districts and within the area of highways is characterized by a high content of heavy metals and is found to be most contaminated. A significant fraction of these harmful chemicals are deposited on the surface of highways and urban roads and eventually transported into local waterbodies by stormwater runoff that is the primary source of water quality impairments in urban areas. This review analyzes and interprets the collected results of tests for the content of heavy metals in sediments from the urban drainage to determine the actual threat they pose to the environment. As a result of the combustion of liquid fuels, significant amounts of nickel, vanadium, cadmium and zinc get into the aquatic environment. Due to the amount of sediments accumulated in the rainwater drainage system, periodic pollution monitoring should be carried out leading to the determination of the method of management and utilization of sludge. The use of separators contributes to the reduction of the presence of heavy metals in the aquatic environment.

1 Introduction

Due to the fast development of industry worldwide the risk of contamination by heavy metals has significantly increased. Pollution of the natural environment by heavy metals contained in water poses a serious ecological problem, especially in industrial areas. Requirements concerning the content of heavy metals in water to be used for drinking are included in [1] which provides the permissible content values (mg/dm^3) for the most toxic heavy metals, that is, cadmium <0.003 copper<2.0 lead <0 [1]. Contamination of water by heavy metals can be of natural and anthropogenic origin. The main source of anthropogenic pollution in industrial areas are production processes. Metallurgic, energetic, mining and chemical industries are one of the biggest sources of heavy metals to be found in stormwater. The basic sources of heavy metals include: industrial and traffic emissions,

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domestic wastewater, agriculture, mining, steelworks, metallurgic industry, landfills, busy road surface runoff [2]. Industrial facilities largely contribute to contamination of water by heavy metals. Operation of industrial plants is accompanied with dumping significant quantities of heavy metals into the water. Transport is one of the major sources of heavy metals emission (wear of tires and roads). The issues connected with occurrence of sediment in a stormwater drainage system have not been explored yet. Not much research on the characteristics of the sediment or its contamination degree has been performed so far. Accumulation of sediment is the effect of sedimentation processes which take place in the stormwater drainage system. Suspensions contain pollutants, that is: heavy metals and persistent organic pollutants. Criteria for assessment of sediment contamination degree have been accepted in order to determine the quality of sediment according to Regulation of the Ministry of the Environment on soil and land quality standards (Regulation of 9.9.2002). Numerous tests indicate occurrence of pollutants in stormwater drainage (that is, heavy metals, chlorides, sulphates, biogenic compounds) adversely affecting the quality of surface water and living organisms. Therefore, it is important to get familiar with the content of the stormwater sediment and provide assessment of its impact on the environment. Contamination by heavy metals reflects the level of air, water and soil pollution by sewage, waste, industrial gases and coal combustion processes. The effects of heavy metal impact appear after many years and they have not been fully explained, yet [3]. Therefore, integrated systems for the Environment Protection leading to reduction or total elimination of emission od substances harmful to the environment should be developed.

2 Material and methods

Data obtained from results of tests carried out in several towns worldwide was used for analysis, Białystok, Belgrade, Beijing, Częstochowa, Guangzhou, Hong Kong, Kielce, Lulea, Lyon, Ontario, United Kingdom and Warsaw are the chosen cities. Data on heavy metal content in sediment from stormwater drainage systems has been analyzed. The results describe samples of sediment collected from street inlets, devices for wastewater treatment such as separators and sedimentation tanks as well as from stormwater storage reservoirs. The tested objects were located in catchments with different characteristics. They differed in terms of vehicle traffic intensity and the catchment management including those with features characteristic of city centers, industrial districts, detached houses, highways and gas stations. The sediment was of diversified character and differed in terms of heavy metals content.

3 Resultas and discussion

Due to civilization development more and more toxic compounds, including heavy metals, can be found in the water environment. Stormwaste water contains big concentrations of zinc and lead, whereas the most contaminated parts of the runoff include: cadmium, copper, nickel and arsenic.

Zinc

Urban stormwater runoff also contains high levels of zinc that impairs the quality of receiving waterbodies. The researchers found a direct correlation between zinc concentrations in urban runoff in sediments from various cities around the world and amount of fuels sold in those cities, which is a surrogate for traffic volume. Urban rainwater runoff contains high level of zinc which has a negative influence on the quality of water courses. Zinc in surface waters exceeds the water quality criteria (120 µg/L) in many urban areas receiving large amount of stormwater runoff [4]. Metallurgic industry, mainly zinc steelworks, are common sources of zinc. Regulation of the Ministry of the Environment [1] provides the maximum zinc content in soil to be (1000 mg/kg). On the example of zinc it can be said that the highest quantity of water pollutants is found in industrial districts.

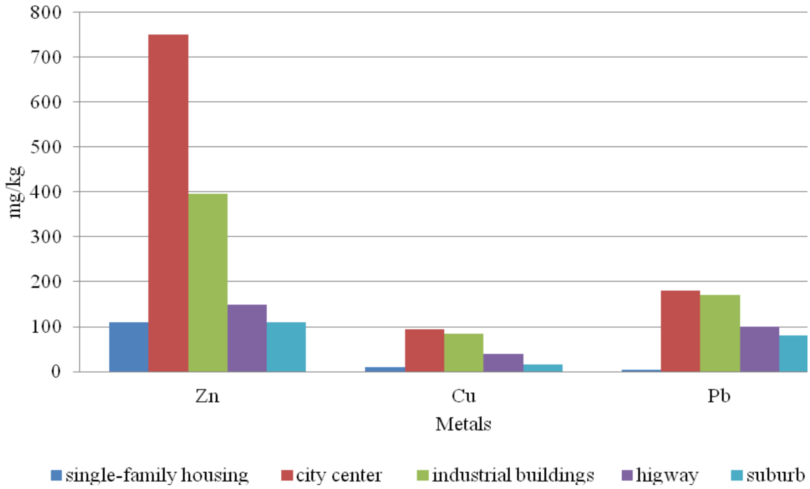

Fig.1. The street inlets, catchment areas of various types.

The highest content of zinc was found in a stormwater inlet situated in a catchment of domestic-industrial wastewater (Fig. 1). Boundary values were exceeded in samples coming from inlets W_{10} and W_{15} in Guangzhou posing a threat to the environment [5]. Boundary values provided in the Regulation of the Minister of the Environment [1] were not exceeded for inlets located in Białystok. The highest concentrations of zinc occur in precipitation wastewater, that is 60-80 %, which is the effect of use of the element in car industry (it is used for coating steel products). The concentration of zinc in sediment from open reservoirs in Calgary was found four times higher than in the most contaminated reservoir '68 St SE" [6]. The highest difference between the samples collected from different sites of an open water reservoir in Kielce, can be observed for zinc whose concentration on the inlet was (70.79 mg/kg), whereas (750.93 mg/kg) on the outlet. This means a ten times increase in concentration [7, 8]. In Great Britain the content of the element in sediment samples was (200.00-2500.00 mg/kg) [9], and in France (640-1809.00 mg/kg). High concentration of zinc exceeded a few times concentration in sediment from reservoirs designed for catchments of domestic type. The concentration of zinc for a catchment of industrial type was a few times higher than that for the domestic type. In the case of sediment tanks and separators the content of zinc in Warszawa was very high (1142 mg/kg) (Fig. 2), [10]. Sediment was collected from devices located at Siekierkowska route and gas station areas where content of this metal was in the range of (859-1195 mg/kg) by Szyprowska. For a sample of sediment collected from the separator at the Siekierkowska route, which is characterized by two roadways (each with three lanes) with an SDR of 100.000 vehicles per day, an exceeding of the concentration of zinc and lead according to the geochemical classification and LAWA is observed. The content of this metal in Canada was much lower (360–290 mg/kg), in India Delhi was (366 mg/kg) (Table 1). Kristin Karlsson described tests of sediment from street inlets in two types of catchments: downtown where Zn was (90 mg/kg) and uptown it was Zn (202 mg/kg). A big number of vehicles and impermeable surfaces contribute to an increase in the amount of Zn (202 mg/kg) in the tested samples those which were collected from downtown street inlets revealed higher content of zinc [11]. The sediments showed a varied content of heavy metals. Large disproportions between sediments for samples taken at inlet and outlet from the reservoir were recorded for zinc (70.8 mg/kg) inlet and (750.9 mg/kg) outlet. The content of zinc in samples from street inlets in China was found to be (2640 mg/kg), (1720 mg/kg) and (915 mg/kg) [5]. The highest quantities occurred in the areas with dense building development and harbor areas and in the proximity of power plants (2640 mg/kg) and (1720 mg/kg). It was caused by vehicle corrosion. Boundary values specified in the respective Regulation of the Ministry of the Environment were not exceeded for inlets located in Białystok. Similarly to zinc deposits from heavy metals, zinc dominates (it accounts for nearly 70% of the sum of all metals).

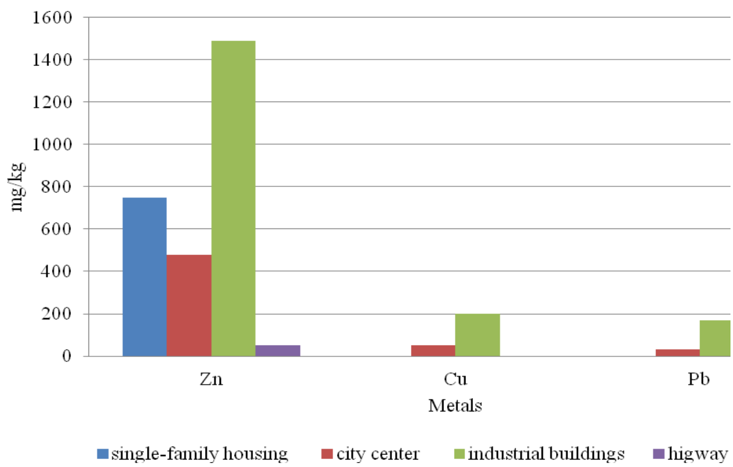

Fig. 2. The average content of heavy metals in sediment samples originating from rainwater retention reservoirs located in the catchment of various types.

Lead

Significant quantities of nickel, vanadium, cadmium and zinc enter the natural environment along with combustion of liquid fuel, diesel and condensed hydro carbonates. The content of lead in samples from z Białystok was significantly lower than the boundary values. Boundary values for lead were lower than for zinc and copper. The content of lead in samples tested in France, from open reservoirs, for the industrial catchment, was a few times higher than for the catchment from residential areas Pb (85.00–323.00 mg/kg) [9] (Fig. 1). In Kielce the level of lead was Pb (0.37 mg/kg) for sediment was taken from a storage reservoir collecting stormwater from a catchment with area 804.6 ha. Situated in an area with housing estates of low-rise buildings and unbuilt areas, open and of rural character, in the south-west part of the town. The content of lead in samples coming from Białystok were found to be much lower than the boundary values. It was observed that only 10% of the discharged lead accumulates on surfaces (at the distance 0-30 m), and the rest is carried over long distances. Significant accumulation of lead in snow was reported. Increased concentration of lead in thaw runoff reached a few (mg/L). The content of lead in Białystok did not exceed 10% of the heavy metal sum in sediment. The highest values of lead content came from a residential area, from a street with the highest road traffic intensity [15]. The content of lead in samples from sediment tanks and separators in Warszawa was (80–858 mg/kg). The highest concentration of lead was reported for Kielce at the level of (0.37 mg/kg). Concentration of the metal in the tested sediment was much lower than the boundary value (200 mg/kg) established for lead, in sediment coming from water reservoirs [1]. In Canada [6] the content of lead was observed to be (6.70–7.30 mg/kg). Tests conducted in China show a diversification in the degree of pollution of sediment from stormwater inlets [5]. A significant increase in lead content was observed in samples from a industrial city of Guangzhou (490 mg/kg), however, boundary values for lead were significantly lower than for zinc and copper. Fuels and products of surface and type rubbing are often the source of lead which is connected with the character of road traffic and its intensity. In samples with water from blocks of power plans the level of lead was (147 mg/kg). In samples coming from an inlet located on a highway leading to an airport (2001 mg/kg) in Lulea. The level of the element was Pb (7.8–20 mg/kg) [11].

Table 1. The annual concentration of heavy metals in selected cities (mg/kg).

Metal	As [mg/kg]	Cr [mg/kg]	Zn [mg/kg]	Pb [mg/kg]	Cd [mg/kg]	Ni [mg/kg]	Cu [mg/kg]	Lit
Poland/Białystok/ street inlets	2.50 –3.00	3.20 –20.20	28.10 –429.00	28.10 –429.00	0.06 –39.90	3.10 –30.60	2.50 –44.60	15
Poland/Częstochowa /urban rainwater runoff	12.10 –28.24	–	–	21.00 –63.00	0.47 –0.77	16.05 –32.12	3.37 –7.99	12
Poland/Kielce/ street inlets	–	0.000 –0.209	0.085 –0.505	0.059 –0.487	0.004 –0.024	0.022 –0.093	0.011 –0.284	8
United Kingdom	–	20.00 –250.00	210.00 –500.00	20.00 –90.00	0.40 –2.20	20.00 –100.00	25.00 –350.00	13
Sweden/Lulea/ street inlets	–	23.00 –32.00	90.00 –202.00	7.80 –20.00	0.04 –0.1	14.00 –17.00	33.00 –39.00	11
Canada/Ontario/ settling tanks and separators	4.00 –9.00	41.00 –74.00	290.00 –360.00	67.00 –73.00	1.40 –1.90	21.00 –26.00	71.00 –98.00	6
France/Lyon	6.60 –13.00	40.00 –77.00	640.00 –809.00	85.00 –323.00	0.40 –8.30	32.00 –54.00	130.00 –349.00	12
China/Guangzhou/ Industrial areas	–	–	129.00 –640.00	70.00 –490.00	–	–	24.00 –201.00	5
Serbia/Belgrade/ urban rainwater runoff	–	0.074 –1.350	0.284 –6.200	0.003 –0.006	–	0.003 –0.010	0.067 –1.820	12
India/Delhi	–	–	366.00	598.00	–	–	721.00	17
Hong Kong	–	–	2902.00	1287.00	–	–	635.00	18
Beijing	–	–	193.00	57.00	–	–	42.00	19

Copper

The content of copper in samples from Białystok and Swedish towns was at a similar level, much lower than the boundary value. Sediment (open reservoirs) was collected from a storage reservoir in Kielce collecting stormwater from a catchment with area of 804.6 ha [3]. The territory covered by housing estates with low-rise buildings and unbuilt open areas of rural character, in the south west part of the city (Fig. 1). Figure 2 present the average content of heavy metals in sediment samples originating from rainwater retention reservoirs located in the catchment of various types. The concentration on the outlet from the storage reservoir in Kielce was about 3 times lower than on the inlet to the reservoir Cu (7.52 mg/kg) [8]. Sediment in open reservoirs in Lyon were characterized by Cu concentration equal to (130.00–349.00 mg/kg). The highest concentration of copper in Kielce for sediment tanks and separators was reported to be at the level of (7.52 mg/kg). The concentration was lower than permitted standards for soil quality and quality standards for land of industrial areas specified on the basis of metals (200–1000 mg/kg) [1]. In Great Britain it was (25.00–350.00 mg/kg) [9], in Canada: (98–71 mg/kg) [6] and in Beijing (42 mg/kg) [19] (Table 1).The highest quantities of copper were found in samples from inlets situated in power plant blocks in China (147 mg/kg) and in Hong Kong (635 mg/kg) [18] and samples from an inlet located on a highway leading to an airport (2001 mg/kg). It was observed that the most common source of copper in sewage sludge was vehicle corrosion. In Lulea the concentration of copper in sediment collected from rainwater inlets was significantly lower: Cu (33–39 mg/kg) [11].

Table 2. Limiting values of pollutant concentrations for the ordinances of the Minister of the Environment according to which classification of sediments from the rainwater drainage system is undertaken (*).

	MEA(*) 16 IV 2002 (Dz.U. 2002 nr 55 pos. 498)	**MEA(*) 9 IX 2002 (Dz.U. 2002 nr 165 pos. 1389)**	**MEA(*) 1 IX 2016 (Dz.U. 2016 nr 0 pos.1395) for 2 group of land**
Metals	Limit value [mg/kg]	Limit value [mg/kg]	Limit value [mg/kg]
Zinc(Zn)	≤1000	300–3000	300–1000
Lead (Pb)	≤200	100–1000	100–500
Copper (Cu)	≤150	150–1000	100–300
Nickel (Ni)	≤75	100–500	100–300
Chromium (Cr)	≤200	150–800	150–500
Cadmium (Cd)	≤7,5	4–20	2–5
Mercury (Hg)	≤1	2–50	2–5

Source: Regulation of the Minister of the Environment

Chlorides

Tests of the average content of chlorides in sediment from devices for runoff sewage treatment in Kielce showed that the highest concentration for samples from a street (4.15–4.9 mg/kg) and a route (6.1 mg/kg). The remaining values ranged between: (0.23–1.63 mg/kg). Sediment from stormwater inlets in Belgrade had similar values Cl: (1.86–8.10 mg/kg) [12].

Cadmium

Cadmium comes from products of type and road surface rubbing; it is used to coat steel objects. The most frequent sources of cadmium are: metallurgic industry, chemical industry, battery manufacturers and municipal wastewater. Concentration of cadmium in France in retention reservoirs is a few times higher for samples from reservoirs of catchments of the residential type Cd (0.40–8.30 mg/kg). In Canada it was about 25 times higher. The content of cadmium in Canada (sediment tanks and separators) was at the level of (1.90–1.40 mg/kg) [6]. In sediment from street inlets in Lulea it was Cd (0.04 mg/kg), whereas in the city center Cd: (0.1 mg/kg) [12]. In Częstochowa the level of cadmium in stormwater runoff was found to be (0.47–0.77 mg/kg) (Table 1). With reference to the cited classification, the exceedance is only observed for the concentration of cadmium in sediments from grooves described by Królikowski et al.

Nickel

Nickel occurs in vehicle fuel and its presence in stormwater runoff is the effect of ground erosion. Concentration of nickel in open reservoirs in Lyon city was found to be (32.00–54.00 mg/kg), whereas in sediment from street inlets in Lulea [11] it was (14–17 mg/kg). In Ontario concentration of nickel in sediment tanks and separators was (21–26 mg/kg).

Arsenic

In the last years emission of arsenic to atmosphere has increased. It comes from combustion of crude oil and black coal. Coal used by large power plants can contain up to (1500 mg/kg). In China the degree of pollution from downtowns and industrial districts is similar due to an industrial character of the analyzed city. Significantly lower quantities of pollutants occur uptowns with detached houses and in the suburbs. In sediment of open reservoirs in Lyon city the content of arsenic was (6.60–13.00 mg/kg) [9,13,14].

Chromium

In the case of open reservoirs in Lyon city the content of chromium was within (40.00-77.00 mg/kg). In sediment of street inlets in Lulea, the level of metal in residential areas was (23 mg/kg) and downtown it was Cr: (32 mg/kg). In Great Britain chromium was observed (20.00–250.00 mg/kg) (Table 1). In Serbia it was Cr (0.074–1.35 mg/kg) [12].

Mercury

The level of mercury in sediment of open reservoirs in Lyon was found to be (0.14–0.82 mg/kg) [9].

4 Conclusions

Sediment from devices for stormwater runoff treatment, especially those located in city centers, industrial districts and within the area of highways is characterized by a high content of heavy metals and is found to be most contaminated. A significant fraction of these harmful chemicals are deposited on the surface of highways and urban roads and eventually transported into local waterbodies by stormwater runoff that is the primary source of water quality impairments in urban areas. Presence of strong relations between pairs of particular metals is connected with occurrence of metals in car industry (eg.: component of metallic varnish, chrome elements). They enter the environment in result of using salt for maintenance of streets in the winter. Due to a lack of adequate rules and norms determining boundary values for chemical contamination in sediment from stormwater drainage systems, it is necessary to encourage introduction of adequate rules to enable regulation of the issues concerning storage and usage of sediment. The analysis of tests results confirms that stormwater runoff from industrial areas pose a real threat to the environment as it brings significant quantities of heavy metals which have a negative influence on the natural environment. Due to phasing out of leaded gasoline, emission of lead from vehicles declined dramatically and has resulted in corresponding decline of lead in road dust and aquatic sediments. It is a good example demonstrating effectiveness of regulations on the improvement of environmental quality. Lead concentrations in road dust and aquatic sediments, however, are still much higher than background levels. Sediment must undergo tests in order to choose an optimal method of its treatment and management. The negative impact of sediment, especially that coming from devices used in severely contaminated catchments, highways, city centers or industrial districts, on the environment must not be neglected. It is necessary to develop and introduce uniform legal laws which will standardize the issues concerning storage and management of stormwater sediment because inappropriate management and usage of devices can lead to contamination of the natural environment and adversely affect the wastewater management system in towns posing a threat to human health and life. This is the method and character of management of a catchment, which uses given devices of the stormwater drainage system, that has the highest impact on the sediment composition. The management method determines the quantity and quality of pollutants accumulated in sediment and indicts possible utilization and management methods. Cyclic monitoring of contamination should be applied in order to identify an select an adequate method for management and utilization of sediment. The highest concentration of heavy metals (Fe, Zn, Cu) comes from pollution produced by motor vehicle traffic. Iron is not considered to be a pollutant but can play a significant role as a medium that isolates and absorbs other pollutants. Sludge formed in rainwater sewers should be subject to environmental testing and classification. It is necessary to create provisions that specifically refer to this type of sludge. The priority should be determining the exact directions for the management of sediments from the rainwater drainage system. The most restrictive of the described classification methods are eco-toxicological criteria defined by PEL and TEL indicators. When determining the content of heavy metals in retention reservoirs, speciation analysis is an indispensable element of the research, allowing to determine their mobility and lability as well as susceptibility to migration from sediments to water depth and conversely.

The results of the tests presented in the study provide the basis for cyclical tests of sediment from urban stormwater drainage system in industrial areas in Bydgoszcz (Poland).

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