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ORIGINAL ARTICLE

The Contamination of Sventoji River Bottom Sediments by Heavy Metals in Ukmerge, Lithuania

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Abstract

Bottom sediment pollution with heavy metals of the Sventoji River in Ukmerge, Lithuania using X-ray fluorescence spectrometry is analyzed in this article. During the research, qualitative and quantitative parameters of heavy metal concentrations and their distribution were investigated. This article presents obtained results of study, where bottom sediment samples were examined from both shores of the river of Sventoji. During this research, received data was treated using GIS software, which helped to interpolate the data of concentrations into the research polygon of the river. GIS software also helped to evaluate the urban runoff influence to the bottom sediment quality and exclude sources of pollution. The runoff dischargers which transport surface wastewater to the river were registered before sampling. At the mouth of streams, flowing into the river of Sventoji, additional samples were taken. After comprehensive river bottom sediment research there is a possibility to assess the extent of anthropogenic activity and its impact on the river ecosystem and human health.

Key words : Heavy metals, River bottom sediments, X-ray fluorescence spectrometry, Concentrations

1. Introduction

The rapid growth of heavy metal concentrations in the environment took place in the first half of the twentieth century because of the industry development. At the end of the twentieth century river sediment studies in Western Europe showed that heavy metal concentrations began to decline (Vink et al., 1999; Middelkoop, 2002). At the end of the Cold War most of the former industrial plants were closed, modern waste water treatment systems started to be installed, those factors may have contributed to the decline of concentrations (Scherer et al., 2003; Zerling et al., 2006) while in the Eastern Europe and Asia the situation remains different. The developing economies are still facing with the environmental contamination of heavy metals and it is still a pressing problem (Gao and Chen, 2012).

Determination of heavy metal concentrations does not help to fully understand the impact of metals for the whole ecosystem or toxicity for each element of the ecosystem, but it is a good indicator of environ-mental pollution (Hooda, 2010). The investigation of heavy metal concentrations in natural environment is often carried out through examining aquatic sediments. Those studies are more accurate comparing to those held on land because the pollution of bottom sediments of lakes, rivers are less likely affected by short term anthropogenic activity.

The river bottom sediments play an important role in water ecosystems, because it transports biogens, creates appropriate environment for aquatic plants

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and hydrobionts. Pollution of river bottom sediments by heavy metals has a negative impact on communities of hydrobionts, can lead to numerous, prevalence changes of species. Metals which are located in sediments can be “locked” in stable fractions, but pH changes can lead them being easily washed out and absorbed by aquatic plants or algae (aquatic sediments can become a source of heavy metal pollution).

One of the major reasons why it is important to investigate the concentrations of heavy metals in sediments of water bodies – they tend to accumulate in food chains and can easily reach the final step – people (Suresh et al., 2012; Taweel et al., 2013; Xiao et al., 2012). Various pollutants accumulated by river sediments are able to migrate to other water bodies. Heavy metals with sediments can be transported to the seas, oceans or lakes also there is a risk that they will contaminate ground water, which is fed by some surface water bodies (Jain and Sharma, 2001; Filgueiras et al., 2002).

Untreated surface run-off from urban areas complements water bodies with heavy metals (Göbel et al., 2007). Aquatic ecosystem bottom sediment contamination with heavy metals can cause the entire ecosystem degradation (Suresh et al., 2012), therefore metal accumulation in sediments raises a great concern for ecologists. That is why heavy metal concentrations in hydro-ecosystems were examined more often around the world (Buggy and Tobin, 2008; Griscom et al., 2000; Karak et al., 2013; Shi et al., 2013; Wang et al., 2012).

There are almost 30 thousand rivers and streams in Lithuania. Those water bodies form Lithuania’s environment and scenic natural landscapes. Rivers and their valleys constitute to ecological compensation areas which neutralize air pollutants, improve microc-limate, stabilize hydrological regime in urban areas and restore biodiversity components (State service for protected areas under the Ministry of Environment, 2012).

The river of Sventoji was chosen for the research. It is one of the longest rivers in Lithuania (246 km). A lot of cities and villages are located on the river banks. Ecological ecosystem balance of river can be disrupted because of anthropogenic activities so it is important to ensure a consistent monitoring of the state of the river. After the investigation of river bottom sediments, determined concentrations should be evaluated using environmental legislation to understand the level of contamination. After doing that, actions can be taken to reduce pollution and the potential negative effects for the environment.

The aim of this study was to assess the contamination of river bottom sediment by heavy metals at Ukmerge city using X-ray fluorescence spectrometry (XRF), compare heavy metal concentrations with environmental legislation and identify the impact of potential pollution sources.

2. Materials and Methods

2.1. Registration of dischargers

Surface runoff dischargers were registered using GPS navigator “Samsung Galaxy Note 3” and “AndroiTS GPS test” application on the right and left river banks in Ukmerge city territory before sampling. The number of dischargers that were registered during the sampling was compared to the official number of dischargers of Environmental Protection Agency of Lithuania. All surface runoff discharges are given in map using GIS software.

2.2. Sampling

The river segment of nine kilometers was chosen for this research. It is flowing through the town of Ukmerge city. This river segment is limited by the highway bridge and the settlement of Leonpolis village. The river segment was divided into equal parts of 500 meters. Additional samples were taken at the inlet streams (Fig. 1). 38 sampling points were arranged on both left and right banks of the river of Sventoji. Sampling started before the highway bridge which crosses the river of Sventoji in Ukmerge city territory. Last samples were collected at the beginning of Leonpolis village.

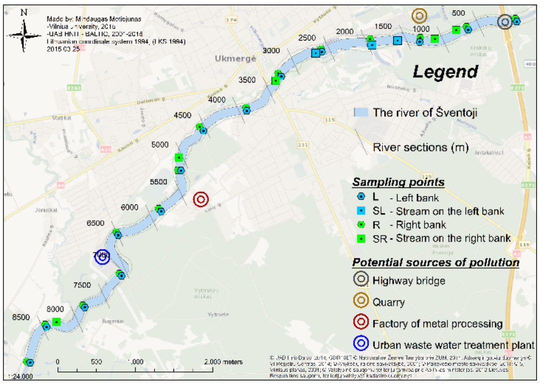

Fig. 1. Sampling points on both banks of the river of Sventoji, Ukmerge.

Samples were taken from the surface (0.10 m of sediment layer, 1 m away from the shore) layer of bottom sediments by swiping 5 cm diameter, 90○ bended plastic tube. Samples were transferred to 100 ml plastic containers to prevent cross-contamination. Samples were recorded, indicating the river side and the sample sequence number.

2.3. Preparation of samples

Samples were transported to the laboratory, where they were placed into Petri dishes and dried in a drying oven for nine hours at 110○ C to the constant weight. Dry samples were filtered through three-chamber sieves – 2 mm, 0.25 mm and 0.125 mm. The obtained filtrate was placed in special caps which were placed into the X-ray fluorescence spectrometer. Samples were analyzed using Thermo Scientific Nitron XL2 Series X-ray fluorescence spectrometer. 28 elements can be detected using this type of fluorescence spectrometer (As, Hg, Cd, Ba, Sb, Sn, Ag, Pd, Zr, Sr, Rb, Pb, Se, Au, Zn, W, Cu, Ni, Co, Fe, Mn, Cr, V, Ti, Sc, Ca, K, S). The further analysis of the results was performed for those metals that are potentially anthropogenic and indicating certain activity impact on the river ecosystem. The total relative analytical error was 5 %.

2.4. Calculation of total contamination index (Z_d)

Arsenic (As), cadmium (Cd), copper (Cu), manganese (Mn), zinc (Zn), zirconium (Zr), vanadium (V) and titanium (Ti) were used in the calculation of Z_d. Background concentrations for those chemical elements are determined in the geochemical atlas of Lithuania (Lithuanian geological survey, 1999).

The extent of pollution in each bank is objectively described by calculating the total contamination index according to the average concentrations of elements in all samples from the each bank of a river. When calculating the total contamination index using average values, the "C" value is the mean value of found element in all samples on each shore. This method gives only one value for the whole shore, so the results can easily be compared.

Total contamination index (Z_d) is calculated according to the formula:

$$Z_d = \sum K_K - (n-1) \quad (1)$$

n – The number of chemical elements;

K_K – The concentration coefficient of chemical element:

$$K_K = C / C_f \quad (2)$$

C – Concentration of chemical element detected in the sample (mg/kg);

C_f – Background concentration of the chemical element (mg/kg);

$^{*}C$ – The average concentration of the chemical element detected in samples of each bank (used for the calculation of Zd by average values).

K_K values are taken into account only if the concentration coefficient is >1. The results are classified into contamination categories: <16 - permissible degree of contamination; 16-32 moderate danger degree; dangerous 32128, >128 particularly dangerous.

3. Results and discussion

XRF method was used to make analysis of 38 bottom sediment samples and identify heavy metal concentrations in the river banks of Sventoji and estuaries of streams. A total of 16 metal concentrations were identified (out of 28 tested) which were higher than detection level. Comparison of the results and the assessment of bottom sediments quality was carried out using the Description of requirements for the management of surface water bodies (Description of requirements, 2014), background concentrations of heavy metals (Lithuanian geological survey, 1999) as well as the calculated Z_d values and interpolation results.

The high degree of variability (Fig. 2.) is typical for the river sediment pollutants (Brown, 2006) which reflects the variety of sources of the contamination within urban catchment and the complex wash-off dynamics of the contaminated materials (Krein and Schorer, 2000; Brown, 2006). Cr, Zn and Cu have the highest variability as these elements clearly reflect the point sources of pollution in the whole river segment that was analysed. Zn and Cu are used to a considerable extent in the technosphere because of their special features. Their applications range from the electrical industry, transport or mechanical engi -neering through to construction and civil enginee -ring. As far as the building sector is concerned, they are traditionally used materials for roof constructions, gutters, drainpipes, chimneys, roof flashings and coverings (Environmental research, 2005). There is also a long tradition of covering entire roofs with these materials.

The largest man-made sources of chromium emissions to the environment are chemical manufacturing, the combustion of fossil fuels, waste incineration, steel making and metal machining. Waste water treatment plants can also be a significant source of high chromium concentrations (Sneddon, 2012).

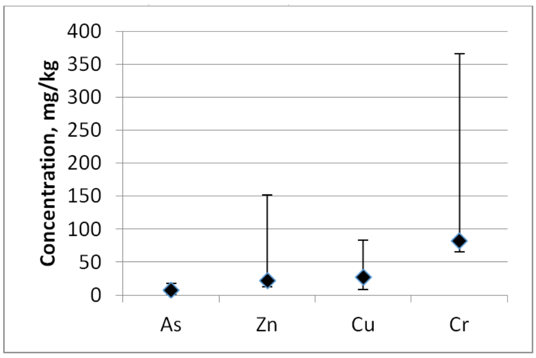

Fig. 2. Maximum, minimum and overall mean concentrations of heavy metals that are potentially of anthropogenic origin.

3.1. Comparison of the results with the maximum allowable concentrations

Average values of concentrations of heavy metals were calculated for both river banks (it was done for those samples, where element concentrations exceeded the level of detection).

Mean values are used for the comparison with the requirements and general qualitative assessment. Results showed, that most of heavy metal concentrations meet the I (first) category of bottom sediments (Pb, Cu, Zn, Hg) according to the Description of requirements for the management of surface water bodies (Description of requirements, 2014).

The concentrations of cadmium (11.89 mg/kg) and nickel (71.5 mg/kg) were only detected on the right bank of river sediments, their concentration meet the II category of bottom sediments (Table 1).

Moreover, relatively high concentration of chromium was measured (177.12 mg/kg) which also meets the II category.

Table 1. Categories of bottom sediments according to the average concentrations of heavy metals

Category	Concentrations of heavy metals, mg/kg (*LOD - level of detection)						
	Pb	Cd	Cr	Cu	Ni	Zn	Hg
I	< 140	< 1,5	< 140	< 75	< 50	< 300	< 1,0
II	140 - 750	1,5 - 20	140 - 400	75 - 1000	50 - 300	300 - 2500	1,0 - 8,0
III	> 750	> 20	> 400	> 1000	> 300	> 2500	> 8,0
Left bank of the river	< LOD*	< LOD*	24,95	22,17	< LOD*	16,92	< LOD*
Right bank of the river	19,61	11,89	177,12	34,72	71,5	29,37	< LOD*

The contamination of bottom sediments by heavy metals is noticeably greater on the right bank comparing to the left one. The concentration of cadmium (Cd) - 8 times, chromium (Cr) - 1.26 times, nickel (Ni) - 1.4 times exceed concentrations required by the I category. Concentrations of heavy metals on the left river bank did not exceed the concentrations that are given for I category.

3.2. Comparison of the results with the background values

To compare results with the background concentration values of heavy metal concentrations in various Lithuanian rivers (Lithuanian geological survey, 1999) it was decided to select only those elements with detection of at least 50% of all samples (Zn; Cu; Mn; V; Ti). Also, the trend of those elements is presented. Assessment of the trend can identify the impact of urban city areas for the river bottom sediment contamination.

Zinc concentrations in bottom sediments of the river >2 times exceeded the background value (58.4 mg/kg) in one sample, taken on the right bank where the stream of "Vilkmergele" flows into the river. The stream flows through the old (center) town territory of Ukmerge city. There are a lot of surface runoff dischargers which are directed to this stream so this clearly shows the urban catchment impact. Zn concentration trend remains constant in all 9 km long segment of the river. It is noted that the city of Ukmerge has a local influence of zinc (Zn) concentration elevations.

Copper (Cu) background concentration value (14.4 mg/kg) was exceeded in 18% of samples taken from both river banks. Trend line showed a decline of Cu concentrations in examined segment of the river. Background concentration of manganese was not exceeded in both river banks. Trend line - remains constant.

Vanadium (V) background concentration value (34.6 mg/kg) was exceeded up to 60% in 24% of samples. Clear differences between the concentration distribution on the left and right bank was not observed. Trend of vanadium concentrations remains stable.

Titanium (Ti) background concentration value (2165 mg/kg) was exceeded in 68% of samples. The highest concentrations were detected in samples taken on the right bank of the river near the highway bridge (background level was exceeded 5.7 times). Despite significantly higher titanium concentrations (comparing to background levels) trend in both banks of a river shows a decrease.

Concentrations of heavy metals in chosen segment of the river of Sventoji decrease or remain stable. Higher concentrations of heavy metals were observed near the highway bridge, city center (old part) on the right bank of the river and at the wastewater treatment plant.

3.3. Results of the total contamination index (Z_d)

Calculation of Z_d is used when the subject (soil,

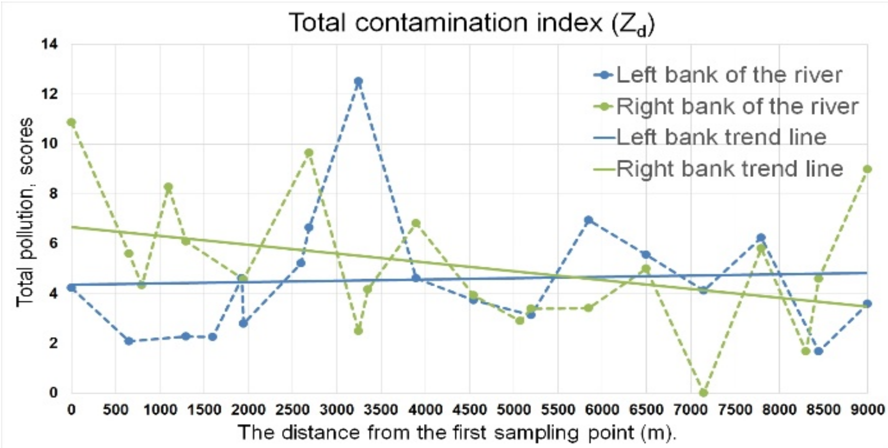

Fig. 3. Total contamination index (Zd) distribution on the left and the right banks of the river of Sventoji bottom sediments.

sediments) is contaminated by several substances or chemical elements (Lithuanian hygiene standard 60:2004).

Z_d helps to identify which bank of a river exposes greater pollution (the average element concentration is used in this calculation). The results of total contamination index calculated for both banks of the river are presented in Fig. 3.

It is noted, that the highest values of contamination are recorded at 3000 m point. This point corresponds to the central part of the city of Ukmerge (Fig. 1). Comparing results with the Description of require -ments for the management of surface water bodies (Description of requirements, 2014) and background concentrations (Lithuanian geological survey, 1999) higher concentrations of heavy metals were more often detected on the right bank of the river. The highest values of Z_d identified in sediment samples meets the permitted limits (<16).

Calculating Z_d using the average values of chemical element concentrations showed that the right bank (Z_d=14.3) of the river is >2 times more polluted than the left (Z_d=14.3) one. Such results are due to the fact that the right bank of the river borders to the central part of the city of Ukmerge. There is more developed infrastructure, were evolving previous factories and now new industries are taking place. The results show an obvious anthropogenic impact on the river ecosystem.

3.4. Results of the interpolation

Values of the total contamination index (Z_d), arsenic (As), zinc (Zn), chromium (Cr) concentrations (mg/kg) were interpolated into the polygon of the studied segment of the river using IDW interpolator in ArcMap program. This method lets visually see the potential sources of pollution, how concentrations vary in different parts of the river.

The results clearly show that there are three main areas where the increased values of Z_d can be observed: highway area at 0 m (Z_d=4-11), central city area at 2000-4000 m (Z_d=10-2.6), the factory area of metal machining and conditioning systems at 5500 m (Z_d=3- 5.5) (Fig. 4).

Increase of arsenic concentrations were detected near the highway bridge (concentration on the right bank is 9.8 mg/kg, >3 times higher than background level), quarry (concentration on the right bank is 6.6 mg/kg, >2 times higher than background level), near the factory of metal machining and conditioning systems (concentration on the left bank is 7.53

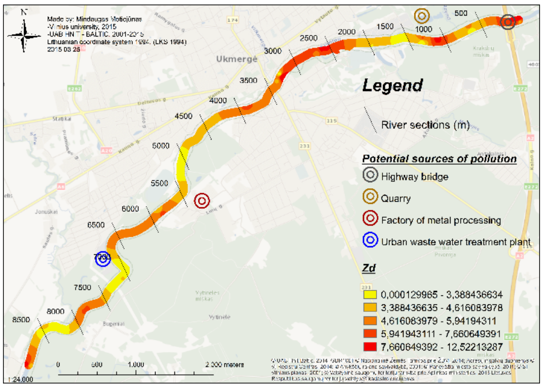

Fig. 4. The distribution of total contamination index (Z_d) in the river of Sventoji bottom sediments at the city of Ukmerge.

Fig. 5. The distribution of arsenic (As) concentrations in the river of Sventoji bottom sediments at the city of Ukmerge.

mg/kg, >2 times higher than background level). Fig. 5 shows the distribution of arsenic concentrations in the study area of the river.

The source of arsenic can be glass, radio electronics (semiconductors), metallurgy industries, thermal power plants and transport. Higher contamination of arsenic was observed in the last century. Arsenic and its compounds were used in pesticides, as well as in rat poison. Arsenic was also used as component in paint. Although inorganic arsenic compounds in agriculture were severely restricted in 1960, it still remains in the soil and water bodies (Bhattacharya et al., 2007).

The highest zinc concentrations were detected near the most densely urbanized area of the city of Ukmerge (2000 – 4000 m point, concentration on the right bank is 129 mg/kg, >2 times higher than back -ground levels). Distribution of zinc concentrations are shown in Fig. 6. The maximum amount of heavy metals enter the rivers from roads, less from the roofs of houses and the smallest amount gets directly with precipitation. The largest amounts of zinc, copper and lead gets from roads, respectively 120, 72 and 62

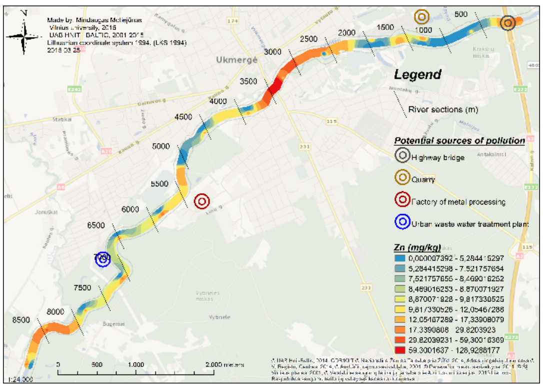

Fig. 6. The distribution of zinc (Zn) concentrations in the river of Sventoji bottom sediments at the city of Ukmerge.

Fig. 7. The distribution of chromium (Cr) concentrations in the river of Sventoji bottom sediments at the city of Ukmerge.

times more than with precipitation (Balevicius et al., 2007).

The highest concentrations of chromium were found near the highway bridge (concentration on the right bank is 230 mg/kg, 4 times higher than background levels) and near the urban waste water treatment plant (concentration on the right bank is 283.6 mg/kg, ~5 times higher than background level). Distribution of chromium concentrations are given in Fig. 7.

Distribution of arsenic, chromium, and zinc concentrations can be directly linked to the potential sources of pollution located in urban area: the highway bridge, most densely urbanized area of the city, factory of metal machining and conditioning systems, and urban waste water treatment plant of Ukmerge.

3.5. Analysis of dischargers

According to the data provided by Environmental Protection Agency of Lithuania there are 5 runoff dischargers on banks of the river of Sventoji. A total of 20 dischargers were observed during this study. Collected results were compared with the Environ-mental Protection Agency data. It was noticed that most of the surface runoff dischargers are not officially registered in the database of Environmental

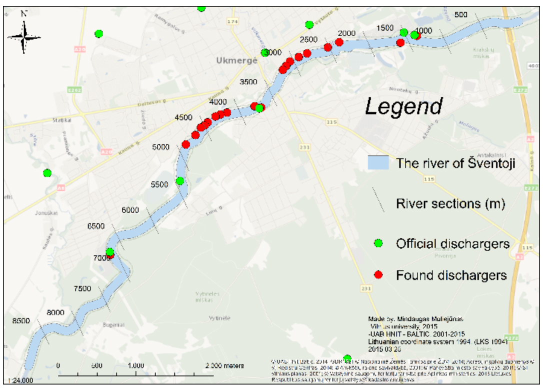

Fig. 8. Comparison of surface runoff dischargers: officially registered by the Environmental Protection Agency of Lithuania and directly observed during this research.

Protection Agency. Almost all unofficial dischargers are located in the central part of a city of Ukmerge. A comparison showed that only 3 points of found dischargers matched with the official dischargers given by the Environmental Protection Agency. Two of the official dischargers were not located during the research (Fig. 8). This analysis shows the real extent of pollution that possibly is getting to the river ecosystem from such number of dischargers.

4. Conclusions

1. The right bank of the river: concentrations of cadmium (11.89 mg/kg) 8 times, nickel (71.5 mg/kg) 1.7 times, chromium (177.12 mg/kg) 1.2 times exceeded the maximum allowable concentrations given for the I category of sediments. The left bank of the river: heavy metal concentrations did not exceed the maximum allowable concentrations.

2. Background value of zinc was exceeded 2 times in 3% of samples tested. Copper up to 3.8 times exceeded background value in 18% of samples. Background value of vanadium was exceeded 1.5 times in 23% of tested samples. Titanium 5.7 times exceeded background value in 68% of samples.

3. The highest total contamination index (Z_d) values were observed near the highway bridge (on the right bank of the river it reaches 10.6) and the central part of the city of Ukmerge (Z_d reaches 9.6 on the right bank and 12.5 on the left bank of the river), but it meets the permitted limits (<16). Z_d calculated by the average values of chemical element concentra-tions showed that the right bank (Z_d=14.3) of the river is >2 times more polluted than the left bank (Z_d=5.8).

5. Interpolation results showed that the total contamination index, arsenic, chromium and zinc concentrations increases near highway bridge, most densely urbanized area of the city, factory of metal machining, conditioning systems and urban waste

water treatment plant of Ukmerge. These results highlight the most dangerous sources of pollution.

6. Comparison of the observed surface runoff dischargers and the official data provided by the Environmental Protection Agency of Lithuania showed that most of the dischargers (75%) were not officially registered in their database. Such results provide valuable information for the future design of monitoring network.

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