Assessment of the effects of urban and industrial development on water and sediment quality of the Drenica River in Kosovo

Fatbardh Gashi[1], Stanislav Frančišković-Bilinski[2]*, Halka Bilinski[2] and Leutrim Kika[1]

[1]Department of Chemistry, Faculty of Natural Sciences, University of Prishtina, M. Teresa 10. 10000 Prishtina. Kosovo
[2]Institute "Ruđer Bošković", Division for marine and environmental research, POB 180, HR-10002 Zagreb, Croatia
*Corresponding author, E-mail: francis@irb.hr

Abstract
Sediment and water samples were taken at eight points along the Drenica River, Kosovo to determine the distribution of metals (Cr, Fe, Ni, Cu, Zn and Cr) in sediments and the water column near potential sources of contamination. The sampling indicated that the largest impact on sediment quality has come from anthropogenic activities, mostly from urban flows, and pollutants from waste water discharges from a nickel refining site. Based on the values of the content of heavy metals in the water and sediment resulting in locations taken in the study, it can be said a nickel refining site is the biggest polluter of Drenica River. Parameters in waters such as electrical conductivity, and total dissolved solids in some sampling points reach relatively high values but do not exceed the maximum concentrations allowed under international standards, even on some places are rather close to them. The concentration of dissolved oxygen in vicinity of the factory is very low and approaches values causing fish kills. According to concentrations of some heavy metals, particularly of Ni, sediments on some stations show values characteristic for highly and extremely contaminated sediments. Further regular monitoring of water and sediment in Drenica River is highly encouraged and advised.

Keywords: Drenica River, water and sediment, statistical analyses, XRF, atomic absorption spectrometry, pollution assessment.

1. Introduction

All over the world the uncontrolled use of natural resources, including inadequate processing of industrial and mine wastes, have caused severe contamination of world ecosystems by heavy metals (Hg, Pb, Cd, Cu, Zn, Ni, Mn) and by a series of organic pollutants (Montgomery, 1996). Sediments (silt and clay) containing toxic heavy metals and other inorganic and organic substances are important for studying water pollution and therefore their multidisciplinary research is essential for understanding different processes and to understand better the geochemical cycles of different trace elements (Halamić et al., 2001). Until recently, the rivers of Kosovo have been poorly investigated. The “State of the Environment in Kosovo 2006-2010, Report” (Prishtina, 2011) prepared by Ministry of Environment and Spatial Planning (MESP) and Regional Environmental Centre (REC) provided the first regional assessment of water quality in Kosovo. The primary goal of the project was to create a database, to assist with the management of Kosovo water resources. Gashi et al. (2009) performed first step with investigation of the rivers Drini i Bardhë, Morava e Binçës, Lepenc and Sitnica, which flow from Kosovo to neighboring countries of Serbia, Macedonia and Albania and therefore are of supra-regional interest. They performed investigations of mineralogical and geochemical composition and of contamination status of stream sediments of mentioned rivers of Kosovo. By comparing the concentrations of toxic elements with the existing criteria for sediment quality, researchers found that two sites in Sitnica River are significantly polluted, especially locations near Fushë Kosova (Kosovo Polje) and Mitrovica. This was assumed to be caused by Zn and Pb processing by flotation and Zn-electrolysis at a nearby factory. In Morava e Binçës River, two sites were found to be polluted with Cd. The authors of that paper suggested future monitoring of sediments and possibly remediation of Sitnica and Morava e Binçës rivers. As Drenica River is the most important tributary of Sitnica River, the current paper presents next step in detailed investigation and monitoring of Sitnica river watershed, which is the most polluted river system in Kosovo. Some other research studies related to Drenica River can be mentioned (Veliu et al., 2008; Krasniqi et al., 2010; Demaku et al., 2012). Measurements were performed in surface and groundwaters, in some springs used for drinking water, and also studying the influence of deposited ash of a nickel refining site, respectively.

Gashi et al. (2011; 2013; 2014) investigated the distribution of heavy metals: Cu(II), Pb(II), Cd(II), Zn(II) and Mn(II) in waters of four main rivers of Kosovo and identified two locations in Sitnica River where sediments were heavily contaminated and would possibly require remediation by Kosovo authorities. Troni et al. (2013) compared the surface water quality in Kosovo in Lumbardhi River basin in the region of Peja. From chemical aspects were investigated some main indicators of pollution as: pH value (in situ), dissolved oxygen, lead, cadmium, copper, zinc, arsenic, cobalt, nickel, uranium, bromine, nitrites, etc. Based on Croatian standards for drinking water, the Lumbardhi River water was classifed in first and second class according to the concentrations of zinc and cadmium. It was classified in the second, third and fourth class according to the concentrations of copper and lead. It was concluded that water resources of Kosovo are endangered by anthropogenic pollution (Faiku et al., 2015).

The aim of the present work is in accordance with recommendation of mentioned previous studies. It is aimed to characterize in details water and sediments of the Drenica River, the largest tributary of Sitnica River, which is the most polluted river in Kosovo and to present assessment of its water and sediment quality.

2. Study area

The Drenica is a river in Kosovo, a 51 km long left bank tributary to the Sitnica River. It flows entirely within Kosovo and gives its name to the surrounding Drenica region. The Drenica originates from the central section of the Carraleva Mountain, in Drenica region. The river originally flows to the north and receives many streams coming down from the Carraleva and Goleshi mountains. The valley of the river is densely populated, with several large villages (Krojmir, Sedlar, Rusinoc, Baicë, Komoran, Koreticë e ulët and Dobroshec) and a small town of Drenas, one of two regional centers of Drenica region. The Drenica River belongs to the Black Sea drainage basin, drains an area of 447 km² and it is not navigable. According to land cover map, issued by European Environment Agency (available at: http://www.eea.europa.eu/data-and-maps/figures/land-cover-2006-and-changes/kosovo/image_large) most of Drenica River valley is under the cover of arable land and permanent crops, parts are under pastures and mosaics, while edges and slopes of the valley are partly forested land. Smaller parts belong to artificial areas.

The study area with the sampling locations is shown in Figure 1 and the details about all sampling sites are presented in Table 1.

3. Materials and methods

3.1. Sampling and sample preparation

Sampling of water was undertaken on 01 April 2014. At each sampling location, water samples were collected in polyethylene bottles. Before taking final water samples, the bottles were rinsed three times with water to be collected. The sampling locations were chosen at points where pollution was expected, due to closeness of traffic, industry, settlements or combinations of those factors. Water samples were collected for analysis according to recommended procedures (WHO, Guidelines for Drinking-water 2006). Geographic coordinates were measured by GPS device Extras, "GARMIN", 12 channel and locations were well described. Each sampling spot, have been marked by codes S_1, S_2 until S_8.

Preparation of water samples was as following: from each sample a quantity was extracted. Samples were filtrated using filter paper "Selecta" No. 589 (Germany), for measuring of water temperature, pH, Electrical conductivity (EC), Total dissolved solid (TDS) and dissolved oxygen. Water samples were preserved according to standard procedures. Each 100 ml was treated with 5 ml of H_2O_2 solution of concentration 5.2M and again vaporized until dry. Dry remnant was again supplied with redistilled water until 100 ml and stored for atomic absorption spectrophotometry (AAS) determination (Skoog et al. 1992; APHA et al. 1998; Alper et. al. 2003; Dalmacija 2000).

River sediments which were in contact with running water were sampled manually, using a metal bucket of the dimensions 25x8x3cm. Sampling was performed at distance from the river banks to avoid possible contamination from the bank material. About 3-5 kg of sediment was collected at each sampling site to provide enough material of fraction <63 μm for geochemical analysis. The material was transported in plastic bags. Samples were dried in air at temperature 20 – 25º C for three weeks. Coarse material was separated using a sieve of 40 mesh and afterwards with the standard sieve of 63 μm, Fritsch (Germany).

3.2. Analytical and statistical methods

The temperature of water was measured immediately after sampling, using a digital thermometer, model "Quick 63142". Measurements of pH and TDS were performed immediately after sampling, using a pH/ion-meter"CONSORT C830" and a "Hanna Instruments, pH&EC" meter. EC was measured by "HANNA Instrument HI 8424" conductivity meter. The accuracy of electric conductivity measurements was 0.1 μS cm^{-1}, of temperature was 0.1°C and pH value was measured with accuracy of 0.01 pH unit.

Metals in water were analysed using an atomic absorption spectrometer model "Perkin Elmer, AAS Analyst 400, HGA 900". The ISO 6351 procedure was followed. Accuracy of determination was ±10% and limits of detection (LOD) were as following: Ni (0.006 ppm), Cu (0.0015 ppm), Pb (0.015 ppm), Zn (0.0015 ppm), Cd (0.0008 ppm), Fe (0.005 ppm), Mn (0.0015 ppm) and Cr (0.003 ppm).

Concentrations of metals in sediments were determined using an XRF model "Bruker Tracer III-SD Handheld". This quick method is assumed to be applied on-site to obtain qualitative or semiquantitative data that assists decisions on further sampling strategy for assessing soil quality. The higher the efforts for pretreatment used on soil samples, the better analytical results can be expected. ISO 13196:2013 was followed, which specifies the procedure for screening soils and soil-like materials for selected metals when handheld or portable energy-dispersive XRF spectrometers are used. The accuracy of determination was ±3% and limits of detection (LOD) were as following: Cr (8 ppm), Fe (5 ppm), Ni (15 ppm), Cu (6 ppm), Zn (4 ppm) and Pb (3 ppm).

The program Statistica 6.0 (Statsoft, 2001) has been used for all statistical calculations in this work: determination of basic statistical parameters and of anomalies (extremes and outliers).

4. Results and discussion

4.1. Physico-chemical parameters and concentrations of some ions measured in river water

In Table 2 are presented values of water temperature, pH, dissolved oxygen, electrical conductivity (EC), total dissolved solids (TDS) and concentration of some ions determined in river water. In Table 3 are presented basic statistical parameters for the selected 8 parameters. In Figure 2 are presented values of selected four parameters and four metals vs. flow direction in km, so that spatial trends in water quality are easily visible.

The water temperature was lowest (8.7° C) at the station S_1, which is closest to the Drenica River source. This is slightly higher than temperatures (7.3° C) of both Rječina and Kupa karstic sources in Croatia, reported by Frančišković-Bilinski et al. (2013). Downstream from Drenica source temperature gradually increases, with some small oscillations. At the station S_8, near the confluence with Sitnica River, temperature was at highest level reaching 13.2° C. This is the usual behavior of most of rivers sourced from mountainous areas that water gets warmer flowing downstream.

Values of pH were highest at the source of Drenica River (7.08), but this value is much lower than pH values that are typically found in karstic rivers of Croatia (pH up to 8.7) reported by Frančišković-Bilinski et al. (2013), possibly due to the composition of rocks in the area. As mentioned,Croatian rivers are situated in karst, while Drenica River is situated in an area of acid magmatic rocks. In Drenica River pH values decreased from the source downstream, and reached a minimal value of around 6.55 at S_5 and S_6 stations, possibly due to a nickel refining site in Drenas, which is located in the vicinity of those two sampling sites. Further downstream from those stations, the pH increases again and before confluence with Sitnica at station S_8 is 6.92.

At station S_1 near Drenica River source, the concentration of dissolved oxygen was 10.82 ppm, what is similar to the value of Zvir spring in Rijeka, Croatia, which is used for drinking water purpose and only slightly less than at Kupa and Rječina river sources

(Frančišković-Bilinski et al., 2013). Dissolved oxygen concentrations declined significantly downstream in the Drenica River until station S_5, where oxygen levels reached a minimum value of only 2.3ppm. This could be due to wastewater from the Drenas and Komaran town sites. At the next downstream location S_6, the dissolved oxygen value is only slightly higher, while at station S_7 it reaches value of 9.62 ppm. Before the confluence of Drenica with Sitnica River, oxygen concentration dropped again to value of 5.22 ppm. Oxygen is a necessary element to all forms of life. As dissolved oxygen levels in water at stations S_5 and S_6 are below 5.0 ppm, aquatic life there is put under stress. The dissolved oxygen concentration at S_5 station is near to the value which can result in large fish kills.

Measured EC values were relatively high along the whole course of Drenica River. The lowest value of 422 μScm^{-1} was measured at station S_1 near the source. Going downstream values gradually increased to the highest value of 798 μScm^{-1}, which is measured at station S_8 near the confluence of Drenica with Sitnica River. EC values measured in this study are much higher of all values found by Frančišković-Bilinski et al. (2013) in the Kupa and Rječina rivers, Croatia, where values range from 200–250 μScm^{-1}, with only one station with value of EC over 300 μScm^{-1}. It is known that higher values of EC might be sign of anthropogenic environmental pollution. Therefore, measured EC values in Drenica River, especially in its lower course, could indicate the presence of significant anthropogenic influence.

TDS values, which are also a possible sign of anthropogenic influence, are rather high in Drenica River in comparison with studied Croatian rivers Kupa and Rječina (Frančišković-Bilinski et al., 2013). TDS values behaved similarly as EC values: at station S_1 near Drenica River source lowest value of 212 ppm is measured. From there, TDS values gradually increased until station S_8 located near the confluence with Sitnica River, where the value of TDS reached 399 ppm. By contrast, in the studied Croatian rivers, measured TDS values were generally less than 200 ppm. Measured values (see Table 2) of all studied metals (Ni, Zn, Fe, Mn and Cr) in filtered water samples from Drenica River were very low. All concentrations of all studied metals, even those on sites with anthropogenic influence, were lower than maximal allowed concentrations from Croatian standards for drinking water, although concentrations of some metals at sampling stations (e.g. Ni at S_6 and S_7; Cr at S_2) were only just below drinking water guideline values. Concentrations of all studied metals changed irregularly along the course of the Drenica River (i.e., there is no clear upward or downward concentration trend). It is interesting that concentrations of some metals (primarily of Zn) elevated at the Drenica River source. This is possibly due to the natural influence of rock composition and the presence of mineralization in the Carraleva Mountain. Decreases of metal concentrations measured in the water column in some locations are likely to be due the processes of adsorption by clay and oxide minerals, the precipitation of minerals and changes in geologic setting.

Evaluation using two dimensional scatter with boxplots diagrams of measured physico-chemical parameters and metals in water samples was also performed. None of those parameters and metals shows any anomaly and their distribution is rather regular. Assessment of the state of heavy metal pollution of rivers is difficult as concentrations are low and the precision of analytical results is also low. Evaluation of heavy metals concentrations in the water column in rivers must be looked with these reservations in mind.

4.2. Concentrations of metals determined in sediments and their assessment

Results of XRF analysis of 6 metals (Cr, Fe, Ni, Cu, Zn and Pb) are presented in Table 4 All results are in ppm. In Table 5 are presented basic statistical parameters for those 6 metals. From the first view on the results, it is obvious that concentrations of all studied metals are lowest at station S_1 near Drenica River source, while concentrations are highest at location S_6, in the vicinity of a nickel refining site. Going downstream from this location, concentrations of all metals gradually decrease, possibly due to limitations in the downstream transport of sediments from one or more point-sources of contamination near this ferronickel production site.

In Figure 3 are presented values of concentrations of metals determined in sediments, vs. flow direction.

In Table 6 are presented results of determination of anomalies (extremes and outliers) for 6 studied metals. Results showed that all elements other than copper showed extreme values, whereas copper levels could be considered to be an outlier. All of the metal extremes and outlier values were measured in samples from the same sampling station: S_6, which is located close to a nickel refining site. This suggests that there is a strong anthropogenic influence on metal levels in sediments at this location. All the anomalously high metal values are extremely high in comparison with data from other river systems of the broader region. Results will be first compared with data of “cluster 3”, which presents average concentrations of particular elements from Kupa River drainage basin, Croatia, reported by Frančišković-Bilinski (2007).

Also, a comparison with available sediment classification standards will be presented. In Table 7 are presented NIVA – Norwegian (Bratli, 2000) and Canadian (Persaud et al., 1993) standards for sediment classification for 6 studied metals: Cu, Zn, Pb, Ni, Fe and Cr. According to those standards, sediments are classified into 5 categories: Insignificantly polluted (I), Moderately polluted (II), Markedly polluted (III), Severely polluted (IV) and Extremely polluted (V). In this table are presented concentrations to which corresponds each class of sediments. In Table 8 sediments investigated in the current paper are divided into classes (I – V) according to concentrations of each studied metal. Results from the table will be shortly presented and discussed.

Even though Cr did not show any statistical anomaly, according to chromium (Cr) concentrations, sediment at S_1 station belong to class (III), “markedly polluted sediments”, while all other locations belong to class (IV), “severely polluted sediments”.

The measured Cu outlier sediment concentration in the Drenica River was 119.3 ppm, what is about 7 times higher than the average Cu concentration in the Kupa River drainage basin (16.20 ppm). According to copper (Cu) concentrations, all studied sediments (S_1 – S_8) would be classified as class (II), “moderately polluted sediments”.

The Fe extreme concentration value in sediments in the Drenica River is 11.63%, what is about 7 times higher than average Fe concentrations in sediments in the Kupa River drainage basin (1.76%). According to iron (Fe) concentrations, sediments at S_3, S_4, S_5 and S_8 stations belong to class (III), “markedly polluted sediments”, while those on locations S_1, S_2, S_6 and S_7 belong to class (IV), “severely polluted sediments”.

The Zn extreme concentration value in sediments in the Drenica River is 6491.2 ppm, what is about 122 times higher than average Zn concentration in Kupa River drainage basin (53.4 ppm). According to zinc (Zn) concentrations, sediments at S_1, S_3, S_4, S_5 and S_8 stations belong to class (I), “insignificantly polluted sediments”, those at stations S_2 and S_7belong to class (II), “moderately polluted”, while sediment at station S_6 belong to class (IV), “severely polluted sediments”.

The Ni extreme concentration value in sediments in the Drenica River is 6501.8 ppm, what is about 246 times higher than the average Ni concentration in Kupa River drainage basin (26.5 ppm). According to nickel (Ni) concentrations, sediments at S_1, S_2, S_3, S_4 and S_5 stations belong to class (I), “insignificantly polluted sediments”, those at stations S_7 and S_8 belong to class (II), “moderately polluted sediments”, whereas sediment at station S_6 belongs to class (V), “extremely polluted sediments”.

Finally, the Pb extreme concentration value in sediments in the Drenica River is 316.9 ppm, what is about 19 times higher than average Pb concentration in Kupa River drainage basin (16.3 ppm). According to lead (Pb) concentrations, sediments at S_1, S_2, S_3, S_4, S_5 and S_7 stations belong to class (I), “insignificantly polluted sediments”. Sediment at S_8 station belongs to class (II), “moderately polluted sediments”, while sediment at S_6 station stations belongs to class (III), “markedly polluted sediments”.

Even when compared with results of Frančišković-Bilinski et al. (2013), with some elevated metal concentrations on some places on Rječina and upper Kupa rivers, maximal concentrations of all metals except Pb are much higher in Drenica River than in mentioned rivers. Concentrations of Pb, Zn, Ni and Fe are also higher in sediments of Drenica River compared to Albanian rivers Mati, Ishmi, Shkumbin and Semani (Çullaj et al., 2005).

As a conclusion of sediment classification evaluation of sediments from Drenica River, it is obvious that for majority of metals upper course of the river belongs to less polluted environment, while sediments from lower course belong to more polluted. Worse situation is at S_6 location, situated just downstream from wastewater outflow of a nickel refining site, where Ni shows concentrations characteristic for extremely polluted sediments. From studied metals Cr and Fe show higher degree of pollution, while all other metals show lower degree of pollution, except on station S_6 where they also have high concentrations.

5. Conclusions

The degree of water and sediments pollution from eight sampling points in the Drenica River was determined in an investigation undertaken in 2014.

All concentrations of all studied metals in water were lower than maximal allowed concentrations from Croatian standards for drinking water. Even where concentrations of metals were low, further monitoring is recommended, due to the fact that concentrations of some metals on some stations (e.g. Ni at S_6; Cr at S_2) were near regulatory limits. Parameters such as electrical conductivity (798.0 Scm^{-1}), dissolved oxygen (9.62 ppm), total soluble substances (399.0 ppm) in some sampling points reached relatively high values, but do not exceed the maximum rates allowed under international standards.

Sediment quality in the Drenica River has been and is currently under the influence of various factors. The main impact comes from anthropogenic activities, mainly from urban flows, pollutants from wastewater discharges, as well as infiltration from agriculture and emissions from a nickel refining site. Based on the values of the content of heavy metals in the water and sediment resulting in locations taken in the study, it can be said that a nickel refining plant is the main polluter of Drenica River.

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Figure captions

Figure 1. Study area with positions of sampling stations and pollution sources.

Figure 2. Downstream distribution of selected four parameters and four metals in water vs. flow direction in km.

Figure 3. Downstream distribution of six metals in sediments vs. flow direction in km.