Carpathian Journal of Earth and Environmental Sciences, February 2017, Vol. 12, No. 1, p. 301 - 310

STUDYING ENVIRONMENTAL LOAD IN THE RIVER BED SEDIMENTS OF BERETTYÓ RIVER AT BERETTYÓÚJFALU, EAST HUNGARY

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Abstract: Signs of anthropogenic environmental load are detected in the river bed sediments of Berettyó River in the section at Berettyóújfalu. The geology of the river bed exposed by boreholes is described and sediment conditions and micro-element compositions determined at every 5cm in the river bed sediments are compared. Results of element analysis were classified based on geoaccumulation index (I_{geo}) and an environmental protection evaluation was performed using the limit values given in the KvVM-EüM-FVM decree 6/2009 (IV.14.). Sediment bodies observed in the river bed are currently formed by erosion and accumulation. Although their morphology and formation mechanism were identified the time of the start of their formation and the rate of accumulation were not possible to determine. Bed sediments are composed of silty, clayey small grained sand of fossil deposition, recent deposited sediment dominated by coarse – medium grained sand and reworked sediment of recent deposition showing recent and fossil characteristics as well. No significant difference between the median of humus content of the three groups was found, however, greatest standard deviation was found in the humus content values of fossil sediments within the groups. Element concentrations measured in every stratum and their changes are in close correlation with grain-size distribution. Element concentration co-change with the change of particle-size can be detected in the case of several elements. Based on geoaccumulation categorization and comparison with limits of the decree, all elements can be classified into contamination free categories (I_{geo}) not exceeding the limit of decree 6/2009 apart from arsenic and silver.

Keywords: Berettyó River; Hungary; river bed sediments, environmental load

1. INTRODUCTION

Significance of rivers is increasing nowadays as water is gradually becoming one of the most important of strategic values. The quality of water in rivers, however, varies in a wide range as they can be easily polluted with no puffer zones. Rapid pollutions, however, run down rivers in a short period and their self-clearing has great potentials (Fleit & Lakatos, 2003, Nguyen et al., 2009, Szabó et al., 2010).

Traces of past pollution events, however, can be found in the sediments of the river bed and the floodplain. Elemental concentrations may increase in certain sedimentary structures over time even if no pollution occurs. Slow accumulation of toxic elements may present environmental risk therefore information on the element concentration of river bed sediments is increasingly significant (Dennis, 2005).

In order to assess the risk of reducing water quality of rivers with a diverse catchment area in a region characterised by both natural and artificial landscapes, the sediments of a medium sized river flowing across variable Romanian and Hungarian lands are studied.

This paper focuses on detecting anthropogenic load in the river bed sediments of Berettyó River along the section at Berettyóújfalu that is regarded to be the most contaminated. Apart from determining the general conditions of river bed sediments the primary aim of the paper is to assess the environmental state of the river bed by studying element concentrations in its sediments.

2. GENERAL CONDITIONS AND DEVELOPMENT OF THE STUDY AREA

Berettyó River coming from Plopiş Mountains, Romania entered the Great Hungarian Plain at Šalard flew towards the marshes of Nagy-Sárrét, W-NW with huge curves. This low-lying area was once the greatest swamp of the eastern Great Hungarian Plain supplied with water by the Berettyó in the whole year. Leaving the Nagy-Sárrét and turning south the river reached its final receiver, the Körös River in the vicinity of the town Mezőtúr. This state was changed fundamentally by river regulation works started in the middle of the 19th century.

The river was diverted into a 14.4 km long artificial canal from Šalard to Kismarja. It was regulated by cutting 44 curves in the section between Kismarja and Bakonszeg. The rivermouth was changed completely as well since a 20 km long new bed was constructed along the former floodplain of the Körös River between Bakonszeg and Szeghalom by 1855 (Fig. 1). As a result, a 106 km long section of the Berettyó was cut away from the new canal of the river diverting it away from the swamp of the Nagy-Sárrét and its mouth into the Sebes-Körös River is located currently at Szeghalom (Ihrig, 1973, Somogyi, 2000). As a result of these regulation works the total length of the river decreased from 364 km to 204 km. Consequently the Berettyó became a channel receiving inland water with a narrow bed (8-35 m) and floodplain (100-150 m, 300-400 m in places) regulated from bottom. Most canals associated with it (Ér, Kis-Körös, Kálló, Kutas Canal) are located in the central and lower parts of the Hungarian part of the river (Korbély, 1916, 1917). Following the completion of the regulation works in 1865 a number of developments were realized along both the Romanian and Hungarian sections of the river improving not only the embankment but the bed and the floodplain of the river as well (Hecker, 1982).

As a result of these works in the 19th and 20th centuries Berettyó River became the most strongly regulated river in Hungary (Dóka, 1997). Apart from river bed constructions the effects of various industrial contamination sources had to be challenged in the 20th century.

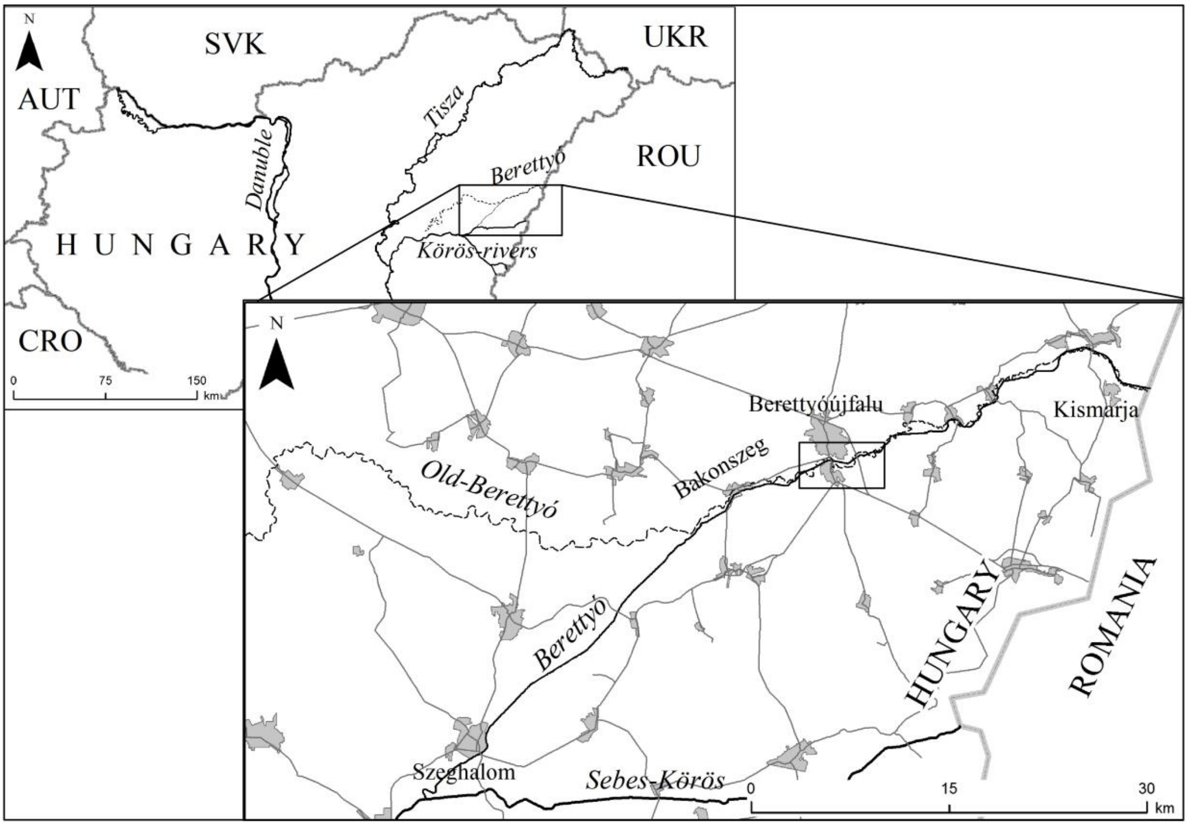

Figure 1. Location of the studied river section

3. SEDIMENTARY CONDITIONS OF BERETTYÓ AND ANTHROPOGENIC LOAD

Sediments accumulated in the alluvium of the river come from the Palaeozoic crystalline schists (staurolite, garnet, biotite containing micaschist, biotite and muscovite containing parametamorphic gneiss, biotitic micaschist, amphibolites, amphibolite schist and phyllite) of the source area and the Palaeozoic, strongly weathered micaschist, gneiss and green amphibolite rocks reworked by streams running down the northeastern side of Plopiş Mountains.

Knowing the main mineral constituents of the bed sediments is important in order to understand the mineralogical and petrographic conditions of the catchment area (Molnár, 1964, Gedeon-Rajetzky, 1973, Szabados, 2002) and to estimate the natural background concentrations resulted by solution from the natural source rocks. Knowing the basic mineralogical composition, the dissolution and related changes of the following elements can be assumed: Fe, Al, K, Mg, Ca (Földváriné, 1975, Balogh, 1991). The character of the natural background concentrations is worth noting in the case of sediments in which extra load of heavy metals is basically well known (e.g. Berettyóújfalu).

Nowadays the entire length of the Berettyó River is exposed continuously to the effects of diffusive and point like contamination sources. Registered contaminations along the Ér Canal and Berettyó River in the last decades mostly originated from outside the Hungarian border. 98% of these contaminations were originated from hydrocarbons. It is promising, however, that the annual frequency of such hazards showed a decreasing tendency in the last three decades (Pádár & Juhász, 1994).

The basin around Suplacu de Barcău lying on the Romanian side of the border is the most industrial area of the catchment of Berettyó. Industry along the Berettyó and its tributaries (e.g. Bisztra) include oil industry, glass-works, mining (Magyari, 1994). Other publications also mentioned the critical environmental state of the river at Suplacu de Barcău. One report said that hydrocarbon contamination in an 8 km long section between Suplacu de Barcău and Blac was so immense that it formed an ecological barrier between the lower and upper sections of the river blocking the movement of species and genes (Andrikovics et al., 2001).

In the Hungarian part the 2678 km^2 catchment area is less industrial, therefore only one industrial contamination source at Berettyóújfalu and several agricultural contamination sources are known (Rácz et al., 2010). Sewage from the Elzett works at Berettyóújfalu established in 1974 can be a significant source of pollution, however, the production capacity of the works decreased continuously from the beginning of the 1990s. Based on the industrial and sewage treatment technologies of the metallurgical works that was split into several smaller companies without a legal successor (galvanization, zinc plating, nickel-chromium plating, electrolytic oxidation, browning, degreasing, phosphate treatment) releasing of the following elements can be assumed: Fe, Cu, Zn, Al, Sn, Pb, Ni, Cr (Orgován, 1989).

Self-purification of the river results in for example that despite the Berettyó is classified as second class or even degraded water (Magyari, 1994) outside classes in its upper sections it is classified as moderately contaminated at its mouth in Hungary (Pádár & Juhász, 1994).

3.1. Interpretation of element concentrations reflecting the sedimentology of the river bed

Anthropogenic load mostly enters waters in dissolved form or bonded to colloids (Winkelmann-Oei et al., 2001). In the clayey, silty bed sediments contaminants bounded to colloids (e.g. heavy metals) may be accumulated in concentrations above the limit in the low energy sections of the river (Waijandt & Bancsi, 1989) while in coarser grained, sandy sediments their accumulation is less probable (Kiss & Sipos 2001). Sediments in the bed and floodplain of the river accumulated over events of several years could present potential risk. This risk is supported by the results of former research carried out on the alluvial samples along rivers Upper Tisza and Lăpus (Csedreki et al., 2011, Gosztonyi et al., 2011, Dorotan et al., 2015).

Sediment bodies accumulating in the active bed cannot be regarded stable forms (Balogh, 1991) since flow and energy conditions in the bed fluctuate rhythmically (Bridge, 1993). Furthermore, the advancement of bed sediment formation either expansional or translational results in different grain-size distribution in the vertical and horizontal composition of the sediment body. As a result, grain structural difference between lower and upper reef complexes will be significant and slight regarding reef development associated with lateral accretion and translational reef development downstream respectively (Miall, 1996).

4. MATERIAL AND METHODS

4.1. Field sampling

Field measurements and sampling were

performed in August 2014. In order to identify the location of bed boreholes, cross sections were drawn at low-water using a Garmint Fishing ultrasound fish radar and a Stonex RTK GPS device. Along the 16 km long section at Berettyóújfalu, five cross sections were identified with 10 boreholes in total. Sampling of the sediments was performed using a 90 cm long Eijkelkamp driller suitable for taking undisturbed core samples. Boreholes were drilled with a variable distance downstream from the section above the town towards the section below the town. In the first three sections (section I, II and III) borehole pairs perpendicular to the river were drilled. In section IV along the longitudinal profile at the sewage supply three boreholes while in section V one borehole were drilled (Fig. 2).

Since the effects of flooding cycles were not always apparent in the structure of the cores taken from the boreholes, their facies interpretation is not part of this paper. Due to the diversity of the sediments exposed by the boreholes, samples were taken from every 5 cm totalling up to 101 samples.

Based on striking changes in the vertical grain-size distribution of the sediment series in the boreholes a boundary was identified between recent and fossil deposits. This boundary, indicated in the borehole sections (Figs. 3, 6, 7 and 8) gives the boundary of recent and fossil depositional environments as well. All sediments deposited in the regulated (from the 19th century) canal of the Berettyó independent of their fresh or reworked character. Fossil sediments include the bed and floodplain deposits of Old Berettyó deposited before regulation works and in which the regulated river has already cut in.

4.2. Exposition of sediments for laboratory analyses, applied limit values

Sediment properties were determined in the laboratory of the Institute of Earth Sciences, University of Debrecen while exposition of the samples and the measurement of their micro-element content were performed at the Measuring Station of Environmental Protection and Nature Conservation of the Hajdú-Bihar County Government Office.

Samples were dried at 40°C followed by sieving (mesh size: 2 mm). Grain-size distribution (Köhn pipette method) and humus content (Tyurin Method) were determined according to the Hungarian Standards in effect (MSZ-08-0205-1978, MSZ-08-0210-1977). Exposition of the samples for element concentration measurements was performed according to the Hungarian Standard MSZ 21470-50:2006 using a THERMO iCAP 6200 type Inductively Coupled Plasma – Optical Emission Spectrometer (ICP-OES) with the help of a central pulverizer (Záray, 2006).

Samples were categorized on the basis of their geoaccumulation index (I_{geo}) determined by Hum & Matschullat (2002) based on their element concentrations. Their environmental state was also evaluated on the basis of limit values declared in attachment 1 of KvVM-EüM-FVM decree 6/2009 (IV. 14). Basis for the six categories of the geoaccumulation index as defined by Hum & Matschullat (2002) is given by element concentration values measured in the clay fraction (<0.002 mm) of the samples taken from boreholes along four sections of the Berettyó River in Hungary.

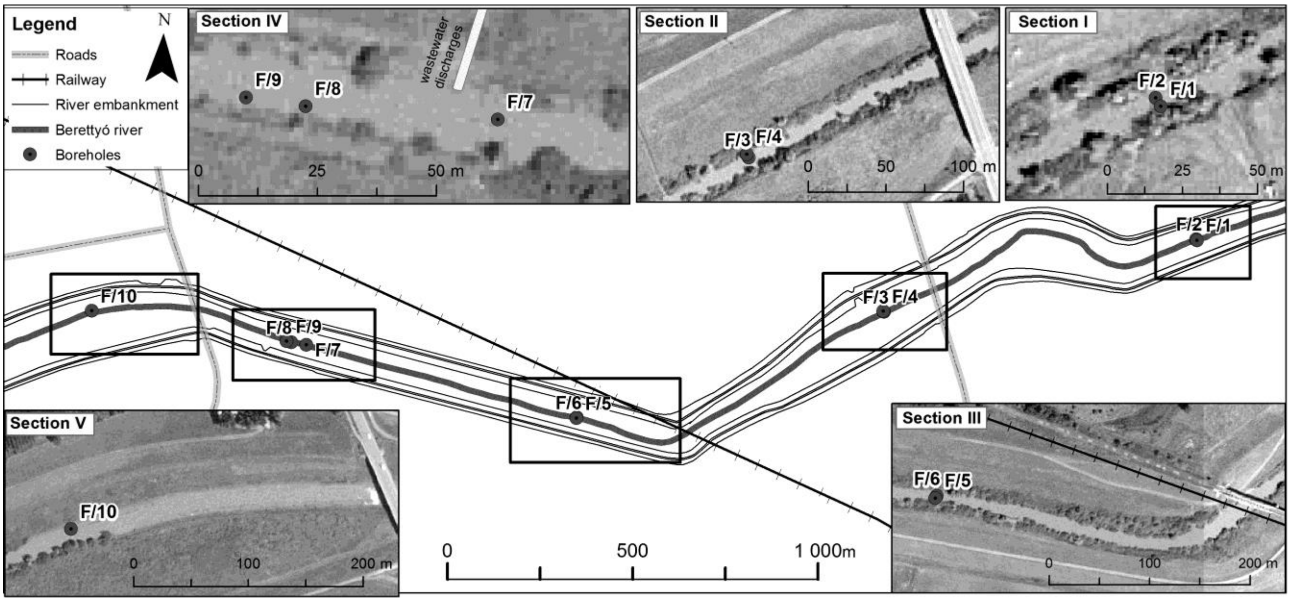

Figure 2. Sampling sites in the research sections identified in the study area

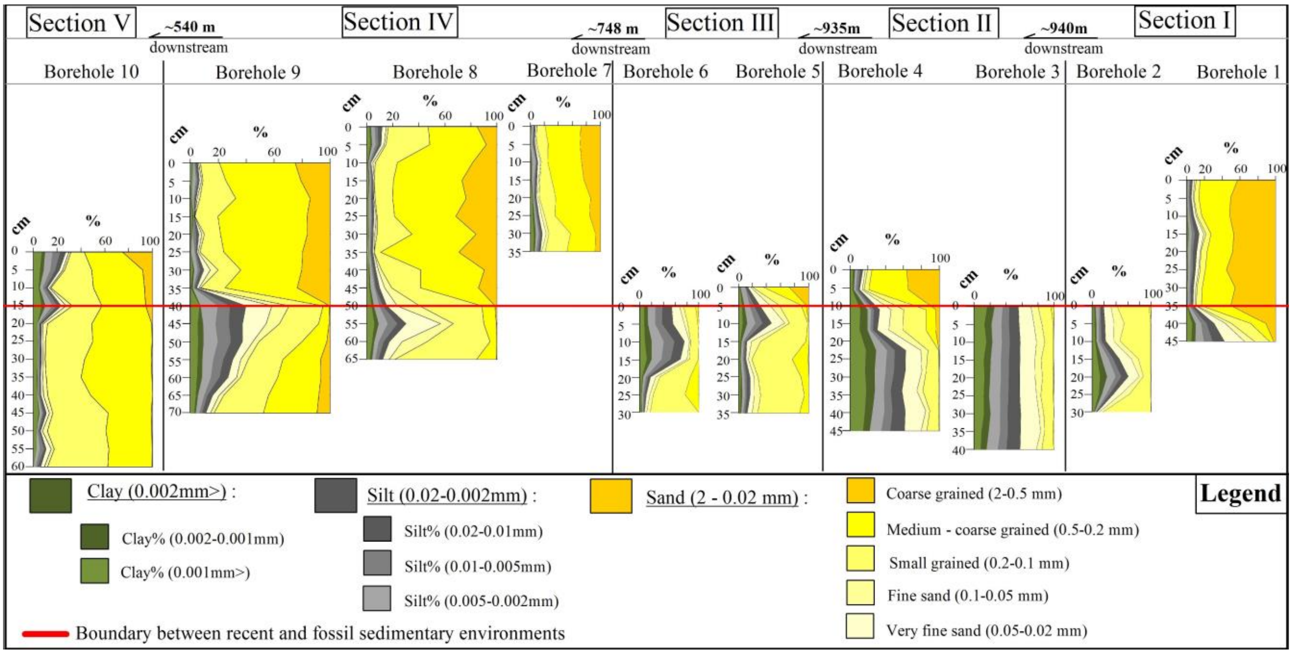

Figure 3. Longitudinal section composed of grain-size charts of boreholes drilled in the studied sections of Berettyó River

Since the ratio of clay in their samples was not published by the authors defining geoaccumulation index, it was not possible to convert it to our samples. Therefore the aim of this research was not measuring geoaccumulation index for our own samples but to use their categories to assess element analysis results.

Graphics of laboratory measurements were performed using MS Excel software, statistical calculations (Shapiro-Wilk and Kruskal-Wallis tests) were performed using Past 3 software while maps were drawn in ArcGIS 10.2 software.

5. RESULTS AND DISCUSSION

5.1. Geological interpretation of the grain-size distribution and humus content of the analysed sediments

Based on their location in the active river bed and accretional topography, the development of sediment bodies is translational. The bottom of the bed in the section at Berettyóújfalu is located 3-3.5 m lower than the surface of the surrounding area. Boreholes have exposed both the recent material of the regulated and the fossil material of the unregulated river. Fine grained fossil sediments in the boreholes can be separated from the coarser grained recent sediments by a sharp change of grain-size. Longitudinal section composed of the grain-size distribution chart of the 10 bed boreholes is divided into ten parts (Fig. 3). Based on lithological observations and the interpretation of the results of laboratory analyses, the boreholes are described in five sections. Description of the sediments exposed by the boreholes in figure 3 and the separation of recent and fossil layers in them are given below according to the sections:

- *In section I* ~40 cm thick recent sediments in borehole 1 are composed of coarse grained and medium-coarse grained sand overlying 10 cm of fossil sediments. The entire borehole 2 is composed of fossil sediments dominated by fine sand and silt.
- In the boreholes of *sections II and III* (boreholes 3, 4, 5, 6) no clear recent layers could be identified probably due to dredging. The sole exception is borehole 4 in which the top ~10 cm is a layer composed of recent sediments. The rest of the material in this borehole and in the others are dominated by fine sand and silt classifying them as fossil deposits.
- *Sections IV and V* are not dredged. The top part of the sediments sampled here (boreholes 7, 8, 9 and 10) are composed of recently deposited but reworked and mixed material with signs of both recent coarse, medium-coarse grained sand and fossil fine sand. In boreholes 8, 9 and 10 recent and fossil deposited layers can be clearly separated.

Fossil layers observed in every borehole have variable composition. Although they are dominated by fine sand the ratio of silt and clay changes according to the appearance of proximal and distal floodplain environment that were characteristic prior to regulation works. Thus fossil layers of boreholes with higher silt and clay ratios (e.g. boreholes 3 and 4) and those with higher fine sand ratio (e.g. borehole 10) were deposited more distant from and closer to the unregulated bed of Berettyó respectively.

Recent layers of the boreholes are dominated by coarser sediments, however, their thickness is variable borehole-by-borehole. These layers can be divided into two groups based on grain-size distribution: one with the dominance of coarse, medium-coarse grained sand and one with material of mixed composition having the signs of both recent and fossil deposits. Both groups, however, were deposited recently. Grain-size distribution of the material of boreholes containing clearly recent sediments (boreholes 1 and 4) is 78-82 % of coarse, medium-coarse grained sand (2-0.2 mm) and 22-18% of finer sand, silt and clay (<0.2 mm). Grain-size distribution of the material of boreholes with mixed layers (boreholes 8, 9 and 10) is 52-78 % of coarse, medium-coarse grained sand (2-0.2 mm) and 48-12 % of finer sand, silt and clay (<0.2 mm).

Fossil, recent and mixed composition of the sediments in the boreholes are clearly illustrated by the ratio of the given grain-size fraction groups. Average ratio of fine sand (0.1-0.2 mm) in fossil layers is 60-80% (except for boreholes 3 and 4) while their ratio in recent layers and in recently deposited layers with mixed composition is around 5% and 15-30% respectively. Similarly the ratios of coarse grained sand (0.5-2 mm) in the three types of sediments are around 0-5%, 38-40% and 17-23%. The box-plot diagram in Figure 4 presents the total humus content of fossil, reworked and recent sediments.

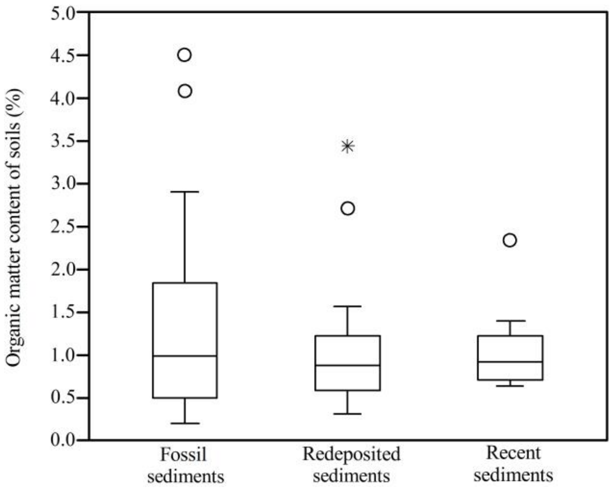

Figure 4. Box-plot diagram of total humus contents (%) in three types of sediments.

As shown in figure 4 the distance between the lower and upper quartile of humus contents is greatest in the case of fossil sediments and smallest in the case of recent sediments. Although the range of humus values is different in the three groups, their median values seem to be almost the same. Since data series totalled by the three types do not show normal distribution according to the Shapiro-Wilk test, the Kruskal-Wallis test was applied considering non-parametric tests to determine the difference among the data series (Table 1).

Table 1: Results of Kruskal-Wallis test

	Fossil	Reworked	Recent
Fossil		0.3441	0.9055
Reworked	0.3441		0.4337
Recent	0.9055	0.4337	

Based on the Kruskal-Wallis test, there is no significant difference ($p>0.05$) among the median of the three sediment types.

Similar humus values of different depositional environments are explained by the similar sedimentation environment of the Berettyó River at any time. As a result, river bed sediments in the studied sections are characterised by general humus content ranges with similar standard deviation and not by extreme humus contents. Exceptions are given by outstanding humus values of layers deposited in sedimentation environments in which the accumulation of the fine grain-size fraction became dominant and vegetation could occur as well (Püspöki & Torma, 2010).

5.2. Results of elemental analysis evaluated from environmental protection point of view

Results of elemental analyses are evaluated according to the fossil and recent character of the sediments and to the grain size pattern of the strata. Special attention was paid to the elements potentially released from technologies applied at potential pollution sources and to the joint occurrence of certain elements in different sediment layers. Analysis of recent and fossil sediments in three boreholes is discussed below.**Borehole 1** is located in sampling section I above Berettyóújfalu. The sampling site is not affected by sewage entry (Fig. 5). Borehole 4 is located in sampling section II. The sampling site is not affected by sewage but anthropogenic load from other sources (e.g. public road, bridge) may appear in the composition of its samples (Fig. 6).

Borehole 9 is located along sampling section IV right after the entry of town sewage (Fig. 7) and is affected by other sources of anthropogenic load (railway and road bridge) as well.

Characteristics of recent sediments

Copper contents are low in the layers of all three boreholes varying between the lower detection limit and 10 mg/kg. Partial concentration co-change with analysed trace elements (Co, Cr, Cu, Ni, Pb) can be observed in the samples of borehole 1.

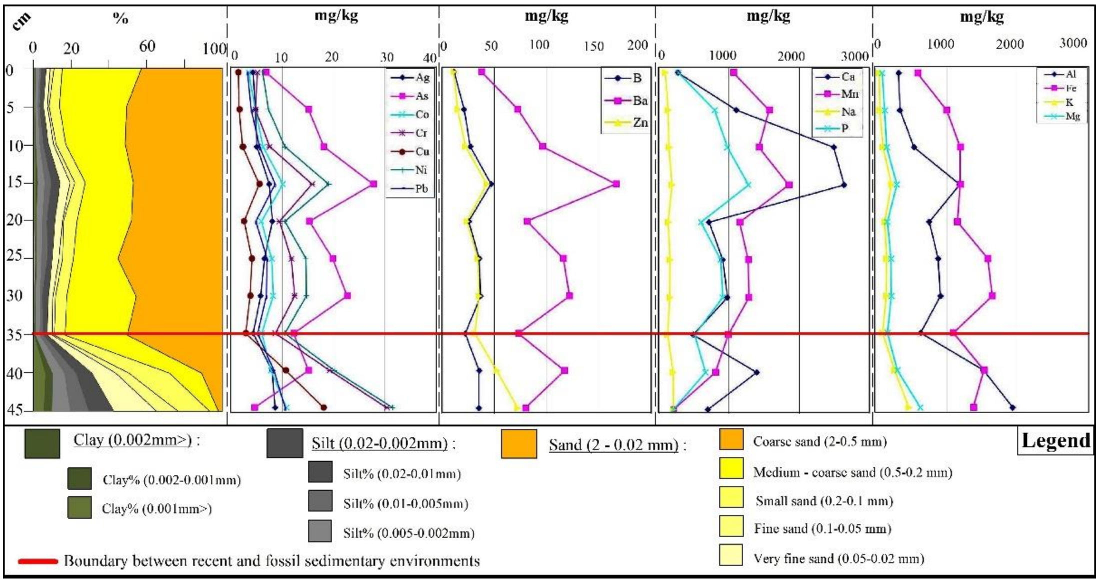

Figure 5. Graphs showing grain size distribution and elemental concentrations of the layers in borehole 1.

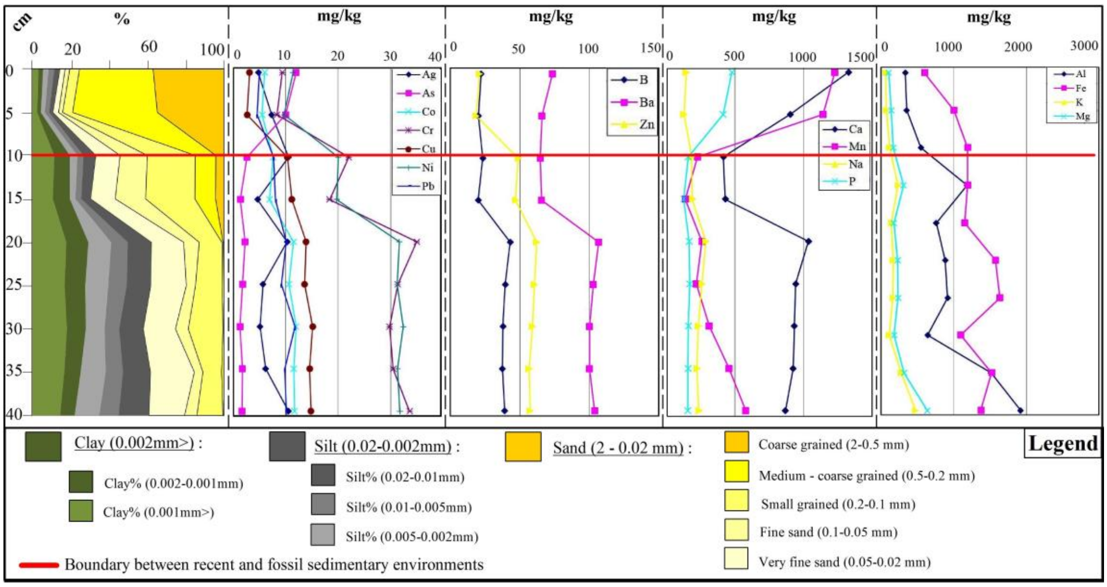

Figure 6. Graphs showing the grain-size distribution and element concentrations of the layers in borehole 4

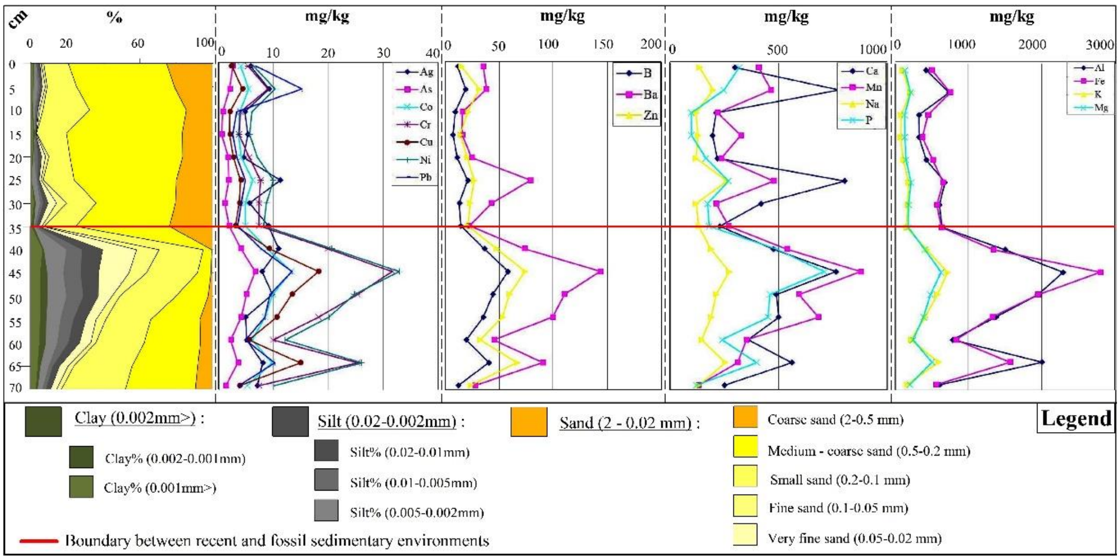

Figure 7. Graphs showing the grain-size distribution and element concentrations of the layers in borehole 9

Silver shows very small concentration values in the layers of all three boreholes ranging from the lower detection limit to 10 mg/kg. Partial co-change with Co, Cr, Cu, Ni, and Pb was observed in the samples of borehole 1.

Arsenic concentrations show decreasing values downstream from concentrations higher than 10mg/kg. It shows partial concentration co-change with Co, Cr, Cu, Ni, and Pb.

Concentrations of **cobalt**, **chromium**, **copper**, **nickel** and **lead** vary around a few 10 mg/kg in the layers of all three boreholes and show characteristic co-change with each other and also with the silt content of the layers.

Concentrations of **boron**, **barium** and **zinc** vary from a few hundred mg/km to several tens mg/km with decreasing concentration values downstream. Barium and zinc show co-change in the layers of borehole 1 while barium and boron change together with each other in the layers of borehole 9.

While **calcium**, **manganese** and **phosphorous** concentrations decrease from a few thousand mg/kg to a few hundred mg/kg downstream, sodium shows no significant change. In borehole 1 P, Ca and Mn, in borehole 4 P and Ca while in borehole 9 P, Ca and Mn change together with micro-elements (Co, Cr, Cu, Ni, Pb) and semi-micro-elements (B, Ba, Zn).

Aluminium and iron concentrations in recent layers decrease from 12-17 thousand mg/kg to 7-8 thousand kg/kg downstream while **potassium and magnesium** concentrations show no significant change. In all three boreholes K and Al show co-change with Mg and Fe respectively.

Characteristics of fossil sediments

Silver contents show very small concentration values in the layers of every borehole ranging from the lower detection limit to 10 mg/kg. Partial co-change with the studied trace elements (Co, Cr, Cu, Ni, Pb) can be detected in borehole 1. Arsenic shows mostly concentration values of 10 kg/kg. Partial co-change with other trace elements in the given layers can be detected in the case of cobalt, chromium, copper, nickel and lead in the fossil layers of boreholes 1 and 9. Concentration of **cobalt**, **chromium**, **copper**, **nickel** and **lead** ranges from several ten mg/kg. Characteristic co-changes could be observed in the samples of all of the boreholes.

Boron and **zinc** concentrations are relatively stable varying around a few ten mg/kg while **barium** changes around a hundred mg/kg. Their concentrations remain stable downstream as well. They show partial and close concentration co-changes with each other in the layers of borehole 1 and 4 and 9 respectively. In borehole 1 Ba and As while in borehole 4 and 9 semi-micro-elements (Ba, B, Zn) show strong concentration co-change with micro-elements (Co, Cr, Cu, Ni, Pb).

Calcium concentrations decrease from one thousand mg/kg to a few hundred mg/kg downstream while **manganese** and **phosphorous** concentrations are greatest in the layers of borehole 1 and smallest in the layers of borehole 4. Sodium concentrations remain relatively stable. Characteristic co-change is detected between P and Mn in borehole 1. Similar co-change is observed among P-Mn-Na-Ca in the fossil layers of borehole 9. In the layers of borehole 1 Ca-Ba-As also show concentration co-change while in borehole 4 Ca and Ba show co-change with the micro-elements (Co, Cr, Cu, Ni, Pb).

Aluminium and iron concentrations vary around a few ten thousand mg/kg while potassium and magnesium range around a few thousand mg/kg. In borehole 1 and in boreholes 4, 9 their partial and close co-change respectively with micro-, semi-micro- and macro-elements can be detected.

Considering environmental protection point of view all measured values of **arsenic** (limit: 15 mg/kg) and **silver** (limit: 2 mg/kg) concentrations in the samples of borehole 1 exceed the limits (6/2009. (IV. 14.) KvVM-EüM-FVM joint decree). In the case of arsenic, high concentrations come from the geological media in the catchment area. While in the case of silver, the limit seems to be too strict.

The presence of known sources of anthropogenic pollution load along both the Romanian and Hungarian sections of the river is a serious problem. Known pollution sources in Hungary are well controlled but there are illegal sources as well the identification of which is not easy. Such industrial, agricultural and residential pollutions are not detected or only in small ratio by environmental protection authorities and mostly only the pollutant material is detected. Such pollutions, however, probably influence the ecological system of the river as well.

6. CONCLUSIONS

Anthropogenic pollution load in the Berettyó River cannot be detected (or only in a slight degree) due to the following geological reasons:

- Recent and fossil units in the sedimentary series can be clearly identified, however, periods of reef development and the age of the sedimentary units were not possible to determine.
- 5 cm thick sections of sampling are too large therefore cyclic strata may result in the homogenization of possible contamination maximums.
- Due to translation mechanisms and assumed

dredging result in the reworking of the studied sedimentary units therefore strata loaded with pollution are washed out and mixed.

- Sedimentary conditions and the micro-element content of fossil river bed sediments are similar in all of the boreholes. Micro-element concentrations measured in fossil layers are several magnitudes higher than that of recent sediments that could be explained by different grain-size and humus content. Thus the dominance of the geological background can be traced in the micro-element concentrations of the fossil sediments.
- The grain-size distribution of the studied strata is in close correlation with their micro-element concentration supporting the assumption that element concentrations are explained by the geological background over anthropogenic influence.
- Analysis of samples taken downstream and upstream of presumed pollution sources yielded no significant difference because the element content of the sediments depends primarily not on their position in relation to the pollution source but on their sedimentary properties mostly grain-size and humus content.
- In the sediments of the studied river section silver and arsenic exceeds regulation limits. This can be explained on the one hand by the relatively low limit of the legislation and on the other hand by the high concentration of silver and arsenic in the upper sections of the river (in Romania) exposed by mining activity.

In conclusion based on the studied sedimentary properties and micro-element concentrations in the sediments along the section of Berettyó River at Berettyóújfalu, the presence of anthropogenic load cannot be excluded but it is subordinate due to the dominance of element concentrations originated from geological sources.

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Received at: 01. 04. 2016
Revised at: 23. 10. 2016
Accepted for publication at: 19. 11. 2016
Publised online at: 24. 11. 2016