

GEOMORPHOLOGY AND SEDIMENTOLOGY OF RIVER BERETTYÓ CONSIDERING ENVIRONMENTAL PROTECTION

József Zsolt PLÁSZTÁN¹, Mihály JÁNÓSZKY², Gábor TALLER³, Zsolt PRÓNAY⁴,
Zoltán PÜSPÖKI⁵, Richard William MCINTOSH⁶ & Csaba Albert TÓTH⁷

¹Department of Physical Geography and Geoinformatics, University of Debrecen, Egyetem tér 1., 4032 Debrecen, Hungary, e-mail: plasztanj@gmail.com

²Synlab Hungary Ltd, Terv u. 92., 9200 Mosonmagyaróvár, Hungary, e-mail: mihaly.janoszky@gmail.com

³Geological and Geophysical Institute of Hungary, Columbus u. 17-23, 1145 Budapest, Hungary, e-mail: taller.gabor@mfgi.hu

⁴Geological and Geophysical Institute of Hungary, Columbus u. 17-23, 1145 Budapest, Hungary, e-mail: pronay.zsolt@mfgi.hu

⁵Geological and Geophysical Institute of Hungary, Columbus u. 17-23, 1145 Budapest, Hungary, e-mail: puspoki.zoltan@mfgi.hu

⁶Department of Mineralogy and Geology, University of Debrecen, Egyetem tér 1., 4032 Debrecen, Hungary, e-mail: mcintosh.richard@science.unideb.hu

⁷Department of Physical Geography and Geoinformatics, University of Debrecen, Egyetem tér 1., 4032 Debrecen, Hungary, e-mail: toth.csaba@science.unideb.hu

Abstract: Riverbed sediments accumulated over recent decades along a 9 km long section of Berettyó River between Esztár and Kismarja were studied from geomorphological, sedimentological and environmental protection points of view. The riverbed along a narrow floodplain was modified several times due to the natural development of bends along the course of the river. Bars developed in the modified environment were sampled with the help of drillcores. Grain size distribution and elemental concentrations were determined at every 5 cm. Ground-penetrating radar measurements were performed as well along the studied section of the riverbed. In order to better understand the radar image and the development of bars three boreholes were drilled into a particular bar.

Based on archive aerial photos, natural and anthropogenic changes in the morphology of the riverbed and in the sedimentation conditions in it can be observed. Forms in the riverbed were exposed by the five boreholes. Based on grain size distribution, the material of the bed can be classified into two groups: fossil material composed of silt, clay and fine sand and a recently deposited material dominated by coarse and medium-coarse sand. Elemental concentrations and their change are in close relationship with grain size distribution. The concentration of several elements changes together. Based on the geoaccumulation classification of the elements and the limit values of the appropriate decree, neither element can be considered pollutants and that neither element exceeded the pollution limit of the KvVM-EüM-FVM decree 6/2009 (IV.14). GPR measurements and boreholes exposed the structure of a 17 m long, ~3-4 m wide and max. 1 m thick sedimentary body. At least four major flooding cycles can be identified in the translational accretion of the point bar based on its graph and GPR profile.

Keywords: riverbed sediments, ground-penetrating radar; environmental load; Hungary

1. INTRODUCTION

Significance of rivers is increasing nowadays as water has become one of the most important strategic values (Áder, 2016). The quality of water in rivers, however, varies greatly as they can be easily polluted with no puffer zones. Rapid

pollutions, however, flow down the rivers in a short period of time and their self-clearing is stronger than one would think (Fleit & Lakatos, 2003; Nguyen et al., 2009; Szabó et al., 2010).

Traces of past pollution events, however, can be detected in the sediments of the riverbed 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 elemental concentrations in riverbed sediments is increasingly significant (Dennis, 2005). Bars may accumulate toxic elements that are released regularly when their sediments are reworked via their migration downstream (Ghinassi et al., 2016). Recently observed increase of flooding frequency may also influence the mobilization and redistribution of older pollutants (Ciszewski & Grygar, 2016).

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 have been studied.

This paper focuses on detecting anthropogenic load in the riverbed sediments of Berettyó River along its section between Kismarja and Esztár. Apart from determining the general conditions of riverbed sediments, the primary aim of the paper is to assess the environmental state of the riverbed by studying elemental concentrations in the sediments of its sedimentary structures.

2. REGIONAL SETTINGS

The Hungarian section of Berettyó River arriving from Plopiş Mountains was completely

transformed during regulation works in the 19th century. The river entering the Great Hungarian Plain was diverted into a 14.4 km long artificial channel at Szalárd until Kismarja. Between Kismarja and Bakonszeg the river was regulated by cutting 44 overdeveloped bends. Even the mouth of the river was changed as a new 20 km long river course was created by 1855 along the former floodplain of the Körös between Bakonszeg and Szeghalom (Korbély, 1916; 1917). With this a 106 km long section of the river was cut and now the river flows into the SebesKörös at Szeghalom avoiding the marshland of the Nagy-Sárrét. Since completing regulation works in 1865 significant investments have helped to improve the floodplain and the embankment system along the river (Ihrig, 1973; Dóka, 1997; Somogyi, 2000).

Aerial photos available since 1951 proved that the channel and floodplain of the river were regularly re-designed in the last few decades. These were justified by the continuous threat of the river leaving its floodplain and by ice congestion (Plásztán et al., 2015). The width of the bed along the 9 km long section of Berettyó River between Esztár and Kismarja (Fig. 1) varies between 10 m and 25 m while that of its floodplain varies between 100 m and 300 m. The current height of the embankment along the studied section was reached in 1963-1965 while the last modification in the route of the bed was made between 1977 and 1978.

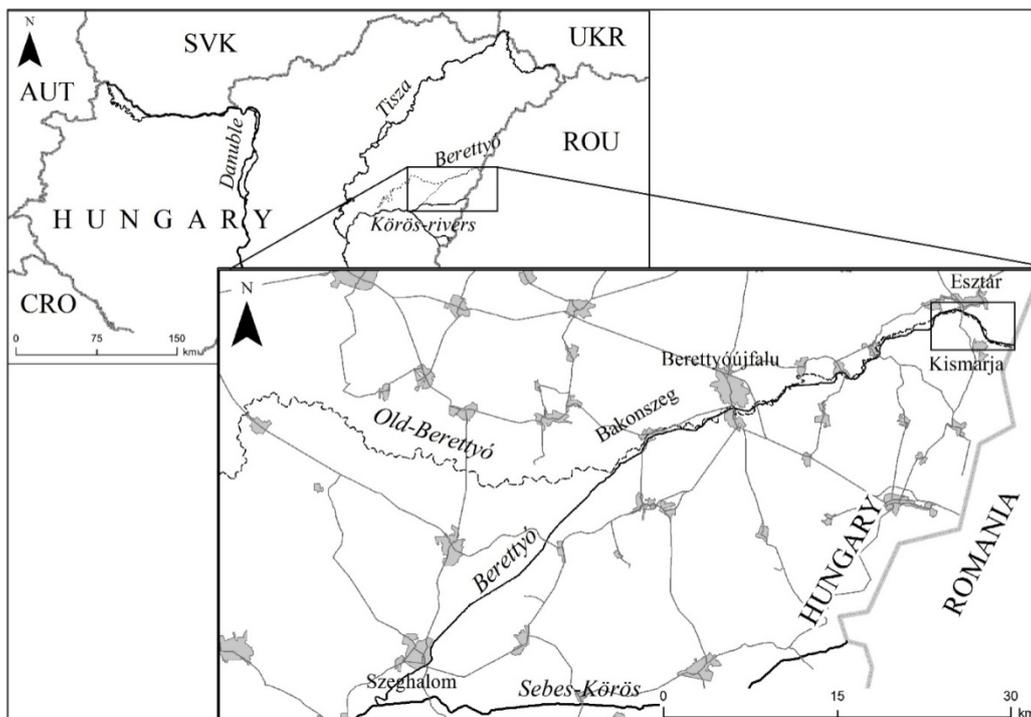


Figure 1. Location of the studied section of Berettyó River

As a reply for intensive anthropogenic impact, the always changing fluvial environment reacts with the formation of bars and the overdevelopment of bends due to the accumulation of sediments and sloping characteristic of the studied section. Sedimentation in this section is very intense resulting in the fast development of sedimentary bodies.

In the last decades registered pollutions along the Ért channel and the Berettyó River originated from outside the Hungarian sections (Magyari, 1994; Pádár & Juhász, 1994; Andrikovics et al., 2001) therefore we were looking for a section to be studied water into which arrives from abroad and represents intense sediment accumulation (Plásztán et al., 2016).

Riverbed or floodplain landforms developed over several years have the potential to store toxic heavy metals and thus represent potential threat on the quality of rivers as suggested by other studies researching alluvial sediments (Csedreki et al., 2011; Gosztonyi et al., 2011; Dorotan et al., 2015).

Understanding the structure and depositional mechanism of the forms studied here is important from the aspect of spreading of pollutants associated with sedimentary grains (primarily the fine fraction). According to the ideas based on classifications according to the qualitative and quantitative models, the interior structure, grain size and stratification characteristics of bars are the result of their expansion or translation dominated development mechanism (Ghinassi & Ielpi, 2015). In the former case, lateral accretion takes place perpendicular to flow direction while in the latter case accretion takes place parallel with flow direction (Bridge, 2003).

The primary aim of the research was to understand the structure and thus development mechanism of bars in the section of Berettyó River between Esztár and Kismarja with the help of boreholes and GPR measurements and to give the environmental protection evaluation of the river section on the basis of elemental concentrations measured in the samples of the boreholes.

3. MATERIAL AND METHODS

3.1. Geoinformatic analysis

Changes in the riverbed over the 55 years (1950-2005) studied on the basis of archive aerial photos were recorded on 13 dates (1951, 1955, 1956, 1963, 1966, 1967, 1971, 1979, 1987, 1989, 1990, 2000, 2005). Photos were taken from Department of Land Survey, Remote Sensing and Land Register of Budapest Capital Government Office (www.fentrol.hu), Department of Environmental

Protection and Nature Conservation, Hajdú-Bihar County Government Office.

Transformation of aerial photos into United National Projection System (EOV) was performed using the software Global Mapper, while adjustment of neighbouring photos and the digitizing of studied morphological changes were performed using the software ArcGIS. The basis for image transformation was presented by geo-corrected MADOP 2005 images. With 8-12 adjustment points on average affine and polynomial transformations were applied.

3.2. Field sampling, laboratory analyses, limit values

Following primary field surveys and the evaluation of archive photo documentation five boreholes were drilled in August 2014 in the bed along the 9 km long section of Berettyó River between Esztár and Kismarja. Sampling was performed using a 90 cm long Eijkelkamp type driller suitable for taking undisturbed core samples. Location of the boreholes was recorded using a Stonex RTK satellite navigation device.

Exposed sediments were separated by 5 cm intervals. The 77 samples taken from the boreholes were dried at 40°C and then sieved using sieves with mesh size of 2 mm. Grain size distribution of the samples was determined based on Köhn's pipette method according to the authoritative Hungarian Standard (MSZ-08-0205-1978). Preparation of the samples and their elemental concentration measurements were made according to Hungarian Standards as well (MSZ 21470-50:2006). Elemental concentration measurements were carried out based on ICP-OES method using a THERMO iCAP 6200 device and central atomizer (Záray, 2006).

Environmental protection based evaluation of the elemental concentrations of the samples was given on the basis of the geoaccumulation index (I_{geo}) of Hum & Matschullat (2002) and the limit values listed in appendix of the KvVM-EüM-FVM joint decree of 6/2009 (IV. 14.).

3.3. GPR survey

Geophysical measurements were carried out with the help of colleagues of the Hungarian Geological and Geophysical institute using a GSSI type georadar in September 2005. Measurements were performed using a 200 MHz mid-frequency antenna. Measurement tracks were recorded using a TrimbleGeoXT satellite navigation device placed next to the GPR in a plastic boat. A ~1300 m long GPR profile was taken between Esztár and Kismarja

(70+100 – 71+400 river kilometres).

Following the interpretation of the GPR profiles (Rashed, 2013) a selected bar was exposed by three boreholes in November 2015 in order to validate the interpretation of the GPR measurements and to understand the structure and development dynamics of the point bar. Sampling was performed using a bottom-valve silt sampler mounted on a PVC pipe. Location of the boreholes was recorded by the TrimbleGeoXT satellite navigation device.

4. RESULTS AND DISCUSSION

4.1. Geomorphology of the studied river section, geological conditions of the riverbed

Resolution of the studied aerial photos was 0.18–2.5 metres/pixel therefore both natural riverbed development and anthropogenic measures could be observed on the photos. Figure 2 presents the studied section with the sections sampled in 2014 are highlighted so that the most striking changes in the main streamline between 1951 and 2005 are also indicated.

Based on the interpretation of aerial photos, most bars developed in the regulated riverbed are mid-channel bars and point bars. The former is formed due to the local widening of the bed and the development of an island in the middle of the bed. The latter type is formed in the freshly straightened

bed diverting the streamline and setting the conditions for forming meanders. Location and development of the bars suggest translational accretion of the sedimentary bodies. The intensity of this mechanism is illustrated by that the length of the bed along the main streamline was increased by 0.5 m / river km between 1951 and 2005 as a result of natural bed development (Plásztán et al., 2016).

Most of the boreholes exposed a bed section formed after anthropogenic bed correction. In this way, the thickness of recent sediments accumulated in the channel-like artificial beds could be studied. Similarly to previous works (e.g. Plásztán et al., 2017) the boundary between recent and fossil sediments in the boreholes was identified on the basis of marked change in the vertical pattern of grain size of the sediments. This boundary between fossil and recent sediments separate not only the deposited material but fossil and recent depositional environments as well. Fossil depositional environments are considered to be the ones in which riverbed and floodplain sediments were deposited from the Ancient Berettyó River prior to river regulation works in the 19th century.

Recent deposition environments have been existing in the bed of the regulated Berettyó River. These are supported by the facts that sediments in the upper ~4 metres in the studied area can be regarded as deposits from Ancient Berettyó (Mike, 1991; Félégházi, 1998) while fossil sediments in the current bed could be sampled from a depth of ~3.5 m.

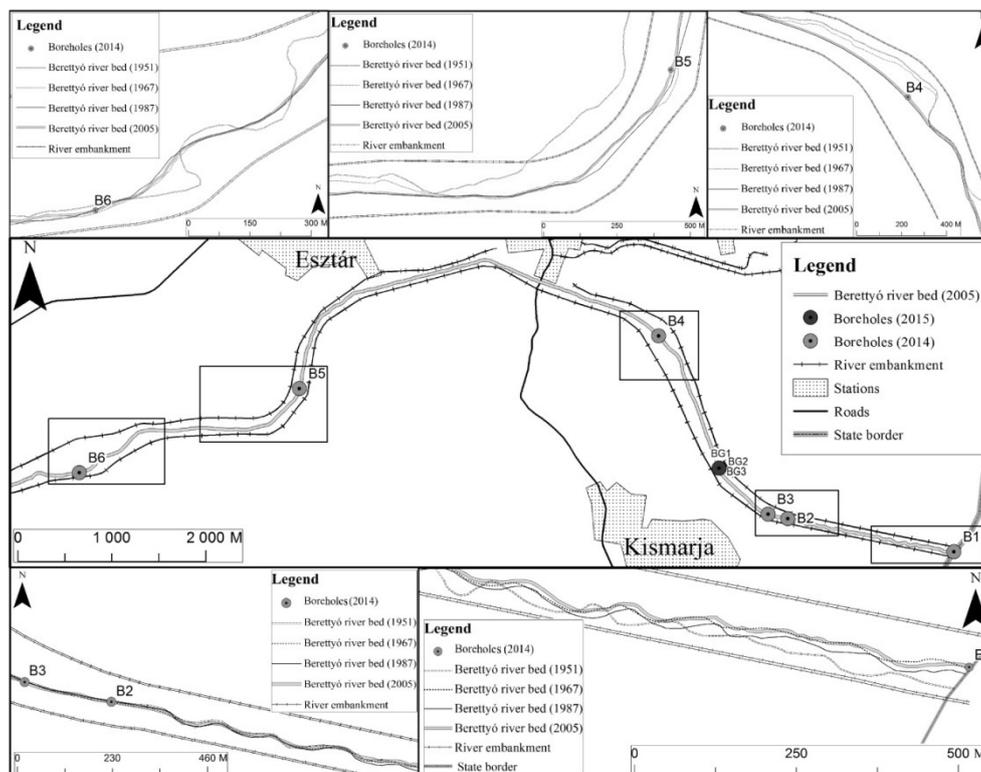


Figure 2. Sampling sites along the studied sections

Figure 3 presents grain size distribution charts of the boreholes in downstream order with the red line showing the boundary between recent and fossil sediments.

Fossil strata identified in all of the boreholes have almost the same composition. Fossil sediments are composed of fine sand (0.2–0.1 mm in diameter) in 20–60%. Together with silt and clay (<0.02 mm) representing 10–55% of the sediments they vary reflecting the distribution of proximal and distal floodplain environments prior to river regulations. I.e. the sediments in boreholes with higher rate of fossil silt and clay (e.g. borehole 1) were deposited further away from the unregulated bed of Ancient Berettyó while the sediments in boreholes richer in fossil fine sand (e.g. borehole 6) were deposited closer to it.

The thickness of **recent** sediments in boreholes varies according to the size of the explored bar and also to which the size of the point bar was explored. Based on the grain size distribution of samples taken from every 5 cm of the boreholes, recent sediments are composed of deposits coarser than that of fossil deposits. Recent sediments in all boreholes can be characterised by the rate of coarse and medium-coarse sand (2.0–0.2 mm) above 70% and rate of finer sand, silt and clay (<0.2 mm) less than 30%.

Recent sediments of boreholes illustrate well

the presence of smaller-sized flow forms in the bed of Berettyó River: ripple marks (boreholes 1, 4 and 6) bars (boreholes 2, 3 and 5). Boreholes exploring bar bodies could not reach the fossil bottom therefore the complete thickness of these bars is not known.

4.2. Development of the landforms in the studied river section on the example of a mid-channel bar (70 + 400 river km)

Sedimentary bodies accumulating in the active riverbed cannot be regarded to be stable forms (Balogh, 1991) because flow conditions along the river undulate rhythmically in almost all cases (Bridge, 1993). It has to be noted that the boreholes exposing the structure of the mid-channel bar and the GPR profile may show differences depending on which side of the form (upper or lower, shoreward or bedward sides) they exposed (Bridge, 2003). The boundary between fluvial landforms and bedrock together with the structure of the fluvial forms can be studied well using a GPR (Best et al., 2003).

Kiss & Sípos (2001) detected similar landforms in Maros River in SE Hungary. They found that one of the most striking river landforms were mid-channel bars. However, they had no opportunity to use GPR profiling to further expose such forms.

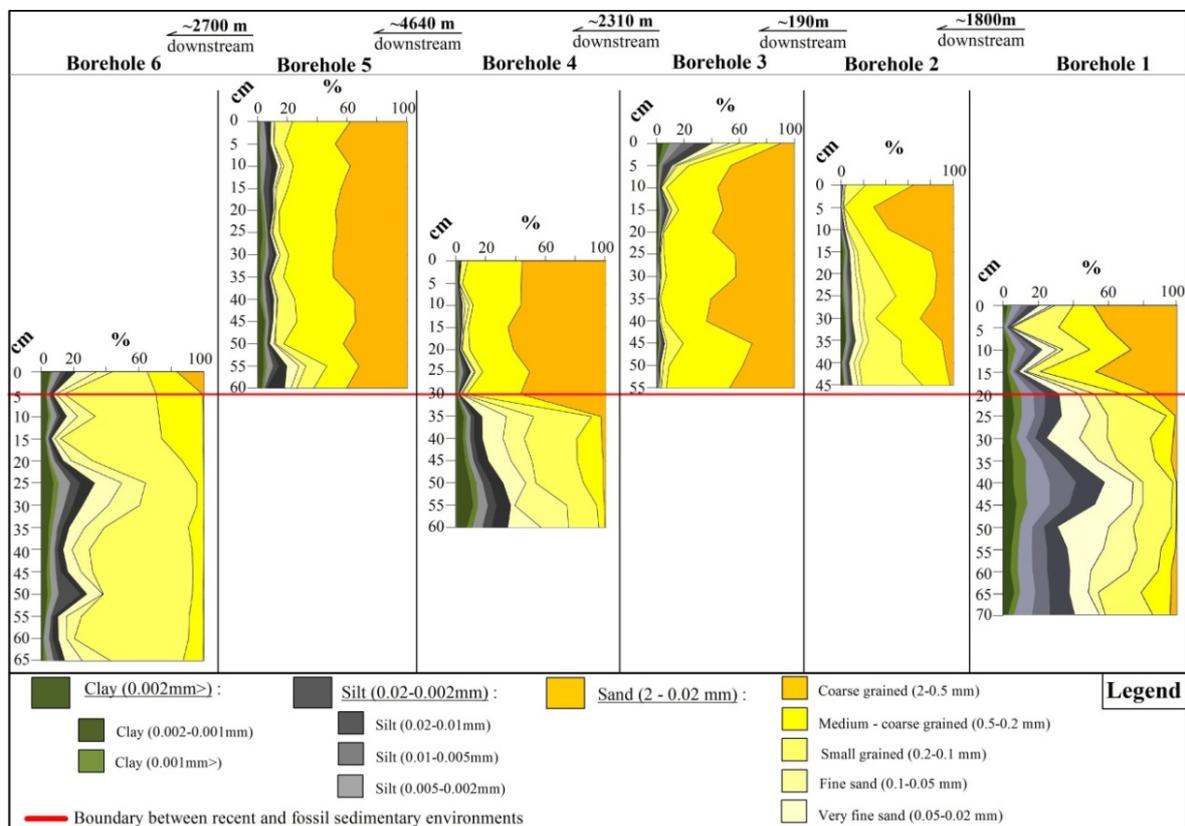


Figure 3. Longitudinal section composed of grain size charts of boreholes drilled in the studied sections of Berettyó River

During the fieldwork GPR profiling and borehole drilling was aimed to measure and expose the composition and structure of the mid-channel bar along its most elevated crest. Based on these measurements, the authors wanted to decide whether the landforms can be explained by expansion accretion or translation accretion mid-channel bar development. This would be important in order to understand the spreading of potential anthropogenic environmental load that could be trapped in the mid-channel bars during sediment accumulation.

Figure 4 shows the radar image divided into periods of the 17 m long, ~3–4 m wide and max. 1 m thick sedimentary body measured by a 200 MHz antenna and exposed by three boreholes (BG1, BG2, BG3) in the riverbed section at 70+400 river km.

Correlation of marked reflection surfaces in the GPR profile and the strata boundaries in the sediments of the boreholes was made possible with the 70% reduction of the height of borehole columns. This difference is caused by the vertical dimension of the GPR profiles calculated on the basis of the propagation of radar waves in water (Taller et al., 2016).

The structure of the mid-channel bar indicates its translational development in the oldest period of which (period I) sediments were deposited directly on the riverbed at the upper end of the form. Onto these sediments that can be regarded as mid-channel bar core – strongly reflecting in the GPR images – coarse sediments were deposited. Following this, the sedimentary body was formed by multiple downstream accretion (periods II, III, IV) and erosion. In the case of the mid-channel bar presented in Figure 4, four periods can be identified three of which were exposed by the boreholes (periods II, III, IV).

Boreholes prove the erosion of upper mid-channel bar parts formed by older accumulation

cycles (periods) depositing finer (10 cm >) sediments of smaller thickness. These thin silty remnants of covering strata with plant remnants appear as marked reflection surfaces on the GPR images (black belts?) separated from the cross stratified sand parts slightly coarsening downward (grey?). Onto the eroded sedimentary body sand sediments and then fine silt were deposited during the following accumulation cycle. The whole mid-channel bar is covered by the loose silty covering sediments of the youngest period widening at the further end (upper part) of the mid-channel bar downstream, at the front of the mid-channel bar (period IV).

4.3. Results of elemental analytical measurements and evaluation from environmental protection point of view

Results of elemental analytical measurements are interpreted according to the fossil and recent origin of the deposits and the pattern of grain size change in the studied strata. Also the joint occurrence of certain elements and co-change of their concentration was studied. In the following the results of the analysis of samples from the strata of three selected, typical boreholes are presented. Regarding their position in the riverbed and their composition, they represent reasonably the sediments in the section at Kismarja of Berettyó River.

Strata in borehole 3 are of recent deposition dominated by coarse and medium-coarse sand (~90-95%). Only the top 5 cm is different where coarse and medium-coarse sand is mixed with silt. Higher concentration of elements in the top layer can be explained by the higher ratio of finer sediment fractions and also by the presumed higher ratio of organic matter content (Fig. 5).

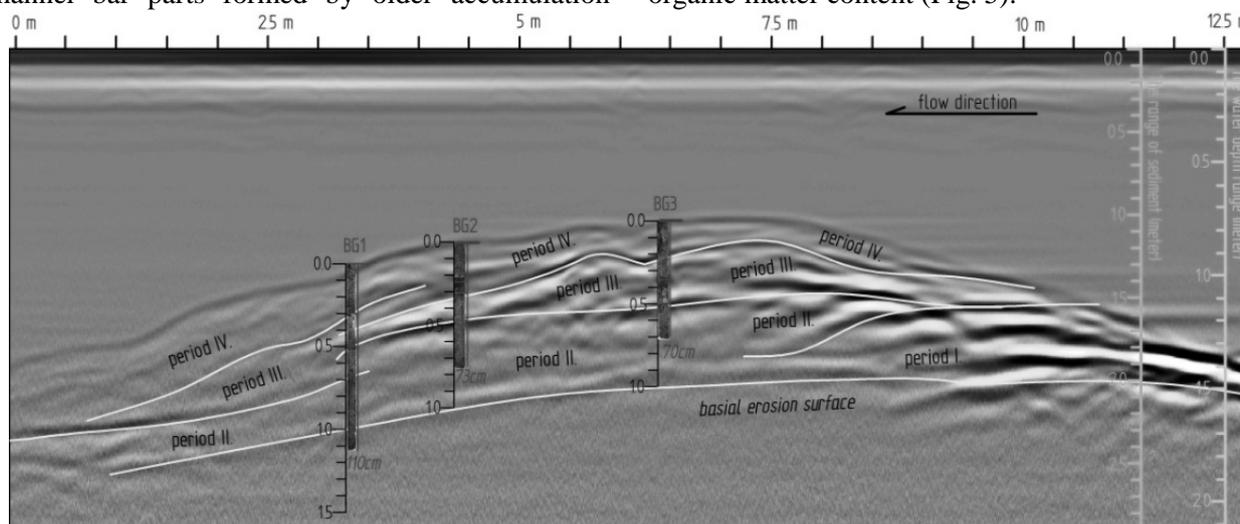


Figure 4. Mid-channel point bar studied on the basis of a GPR profile and three boreholes

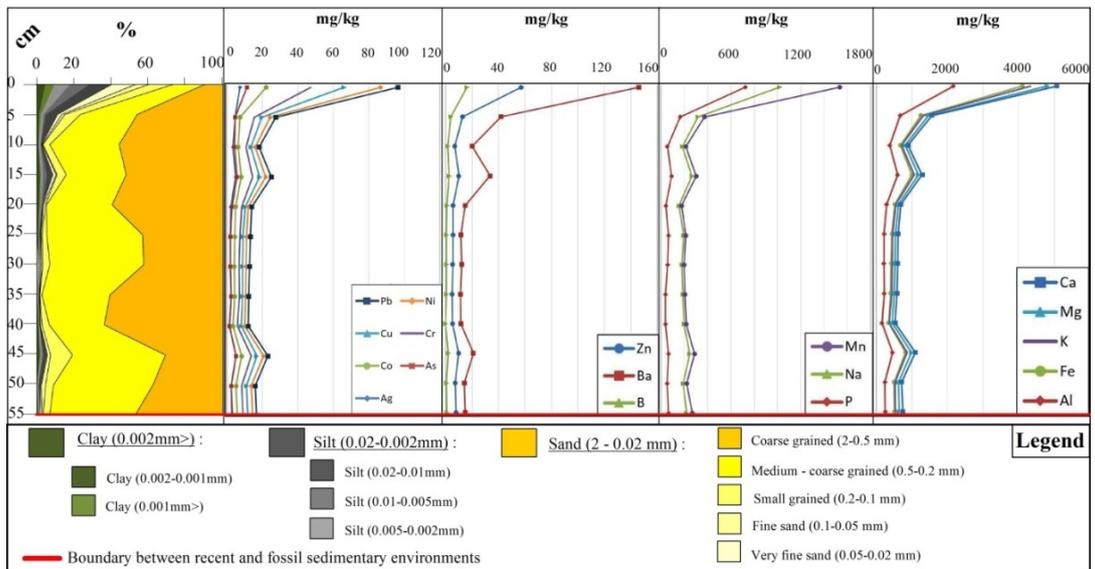


Figure 5. Graphs showing grain size distribution and elemental concentrations of the strata in borehole 3.

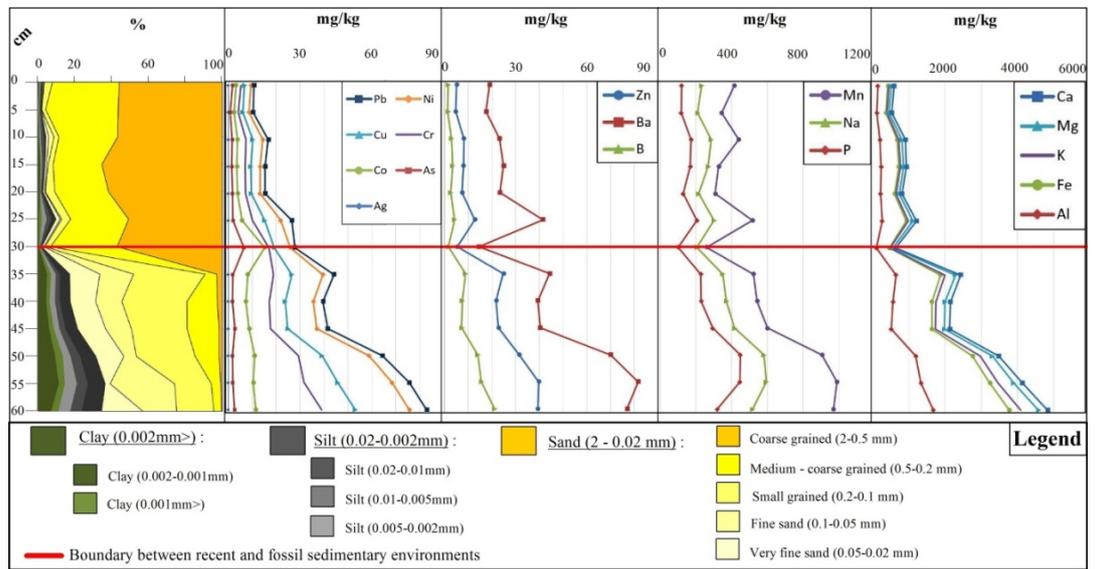


Figure 6. Graphs showing grain size distribution and elemental concentrations of the strata in borehole 4.

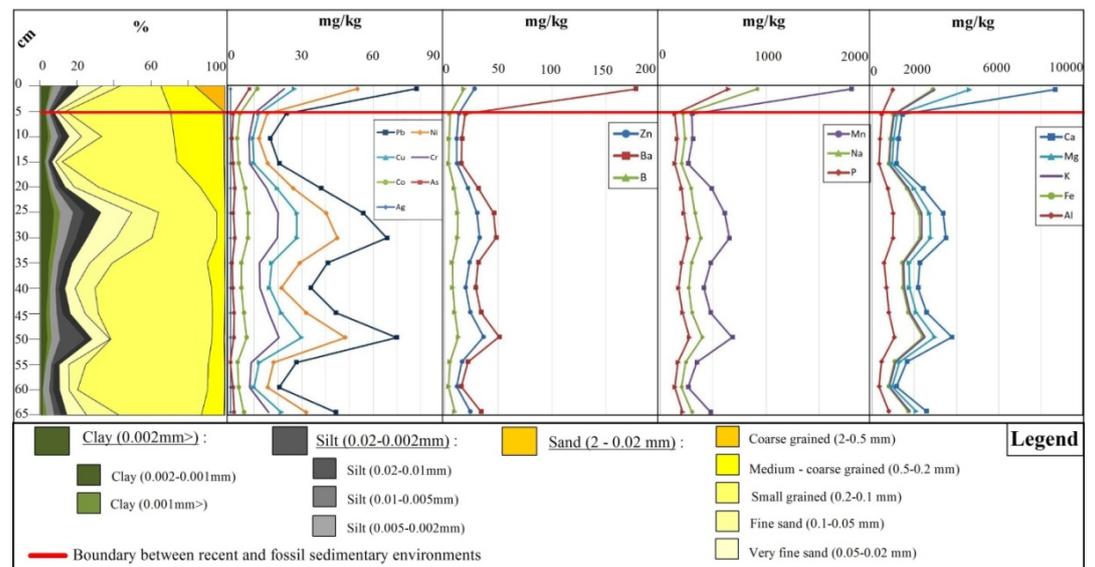


Figure 7. Graphs showing grain size distribution and elemental concentrations of the strata in borehole 6.

Borehole 4 cuts a ~30 cm thick layer of recent sediments with coarse and medium-coarse grain size characteristic for recent deposits. The bottom ~30 cm thick layer of the borehole is composed of fossil sediments. These fossil sediments are characterised by an increase of the ratio of fine sand, the disappearance of coarse sand and a higher ratio of silt (Fig. 6). Borehole 6 contains sediments of fossil origin except for the top 5 cm showing coarse sand dominance characteristic for recent sediments. The remaining 65 cm is dominated by fine sand and silt (Fig. 7) characteristic for fossil sediments.

Concentration of the studied elements is surprisingly high in the recent sediments in the top 5 cm of boreholes 3 and 6 compared to the general values measured in recent sediments. These unusual concentrations of the topmost layers are not included in the following discussion since they are not characteristic for recent sediments. Therefore the discussion of the results is given based on the recent sediments of boreholes 3 and 4 and on the fossil sediments of boreholes 4 and 6.

4.3.1. Characteristics of recent sediments

Silver concentrations are below the lower detection limit except for the strata of borehole 3 in which copper could be detected clearly with concentrations varying around 3 mg/kg. **Arsenic** varied around the lower detection limit in the strata of borehole 3 and showed concentrations below the lower detection limit in those of borehole 4. Concentrations of microelements (Ag, As, Co, Cr, Cu, Ni, Pb) showed values around 10-20 mg/kg.

Boron was found below the lower detection limit, **barium** was found in 20-50 mg/kg while **zinc** was present in a few tens mg/kg. Concentrations of barium and zinc increase downstream.

Phosphorous and **manganese** concentrations ranged from a few tens mg/kg to <200 mg/kg, increasing downstream. **Sodium** was found in the strata in concentrations between 100 and 200 mg/kg.

Typical concentration value for **potassium** was 400 mg/kg, while that of **magnesium** and **calcium** varied around 900 mg/kg in recent sediments. **Aluminium** and **iron** varied rhapsodically and can be characterised by a range between 1500 mg/kg and 7000 mg/kg. Co-change of all measured elements is strong with both each other and the sand fraction.

4.3.2. Characteristics of fossil sediments

Silver and **arsenic** contents were found to be below the lower detection limit. Concentrations of the studied microelements (Co, Cr, Cu, Ni, Pb) vary rather hectically between 3 and 80 mg/kg with

increasing values downstream and downwards in a borehole.

Boron was found in concentrations around the lower detection limit while **zinc** was present in 10–30 mg/kg and **barium** in several tens mg/kg. While concentration of the studied elements (B, Ba, Zn) in the strata of borehole 4 increase with depth it was undulating in the samples from borehole 6.

Manganese, sodium and **phosphorous** contents vary between 100 and 1000 mg/kg and tendencies are similar to those discussed above in the case of boron, zinc and barium.

Aluminium concentrations were <2000 mg/kg in the samples of both boreholes. **Iron, manganese, potassium** and **calcium** were measured to be present in a few ten thousand mg/kg and a few thousand mg/kg in the samples of borehole 4 and 6 respectively.

Co-change of elements measured in fossil strata is close except for silver and arsenic. They also change together with silt and sand fractions.

4.3.3. Elemental concentrations considering environment protection

Elemental concentration values measured in the silt (<0.02 mm) samples taken from four Hungarian sections of the Berettyó River were used for classifying the samples in the six classes of the geoaccumulation index (I_{geo}) of Hum & Matuschullat (2002). I_{geo} classes were determined in the case of the following elements: Fe, Mn, Cd, Zn, Pb, Cu, Cr, Ni, Co, Sb, As. Threshold values are given in % for iron and ppm (mg/kg) for the rest of the elements.

Regarding **iron content**, most of the sediments are classified as *free of pollution – slightly contaminated* ($Fe < 14.10\%$) while certain samples can be classified as *slightly contaminated* ($Fe < 28.20\%$).

Considering **cadmium content** measured in the boreholes, most of the sediments can be classified as *practically free of pollution* ($Cd < 0.45$ mg/kg), while certain layers (layers in the depths of 50–65 m and 70–75 m in borehole F1 together with the layer at 0–5 cm in borehole F3) are classified as *slightly contaminated* ($Cd < 1.8$ mg/kg).

Concentrations of **manganese, zinc, lead, copper, chromium, nickel, cobalt** and **arsenic** measured in the sediments are classified as *practically free of pollution* ($Mn < 1275.0$ mg/kg, $Zn < 142.5$ mg/kg, $Pb < 30.00$ mg/kg, $Cu < 67.50$ mg/kg, $Cr < 135.00$ mg/kg, $Ni < 102.00$ mg/kg, $Co < 28.00$ mg/kg, $As < 19.50$ mg/kg).

Based on the limit values listed in KvVM-EüM-FVM decree 6/2009 (IV. 14.), it can be stated that elemental concentrations exceeding the legal

limit were found only in the samples of borehole 3 (silver, the limit concentration is 2 mg/kg). Maximums of silver concentration can be measured in the silty layers closest to the main body of water.

It is difficult to find similar measurements in the literature related to other rivers in Hungary or in other countries in the Carpathian Basin. Kiss & Sipos (2001) carried out the measurement of some elements in Maros River, SE Hungary. They studied the relationship between grain size and the concentration of Cd, Cu, Pb, Zn and also that of organic matter content and the concentration of the above heavy metals. They obtained results similar to those of the present paper. The concentration of heavy metals increased with decreasing grain size in the sediments of the river and also with decreasing organic matter content. They found also that certain fluvial landforms could act as traps for certain elements and may release these elements at any time increasing the risk of environmental load. The concentration of the studied heavy metals, however, did not reach or exceed legal limits.

It might be possible to identify and separate recent and fossil sediments along the Maros River as well using the methods presented in this paper, however, probably the results would be the same.

In another study Kozák et al., (2002) measured Cd, Co, Cu, Fe and Ni among others in the water and sediments of Eger Stream, NE Hungary and in soil samples from right next to the stream in order to investigate the emitting of the geological environment and a middle-sized town (Eger) with some industrial establishments.

Similar to Berettyó River Kozák et al., (2002) found that the geological formations determine primarily the elemental concentrations measured in the soils near Eger Stream and in the sediments and water of Eger Stream. They also stated that the concentrations of the heavy metals changed together with the grain size of the sediments.

In the present paper not only heavy metals but numerous other elemental concentrations were found to be changing together and with grain size and also with the type of the studied sediments (recent, fossil). Compared to the current legal limits, however, elemental concentrations mostly stay below them similar to the results of Kiss & Sipos (2001) and Kozák et al., (2002).

In a more recent paper Comero et al., (2014) identified three groups of elements that characterise natural and anthropogenic sources in the sediments of Danube River using the positive matrix factorisation (PMF) method. Similar to the Berettyó River, most measured elements (many heavy metals among them) in the sediments of the Danube River

reflect mostly natural rather than anthropogenic sources, however, the co-change of certain elements (Hg, Pb, Zn, Cu, Co and As) suggests an anthropogenic origin (Comero et al., 2014).

5. CONCLUSIONS

Based on archive aerial photos, boreholes and GPR profiles the presence of sedimentary bodies in the river bed along the studied river section was proved and their morphological and structural conditions could be studied.

In all of the boreholes the boundary between recent and fossil sediments could be identified on the basis of the marked change of grain size and elemental concentrations. Concentration of the studied elements follows the pattern of the grain size fraction <0.2 mm.

Geophysical measurements proved that the studied mid-channel bar indicate translational development mechanism based on its position in the bed of the river, accretion topography and sedimentary structure.

Anthropogenic environmental load can be hardly, or cannot be detected at all due to the following geological, sedimentological and sediment quality reasons:

- Grain size distribution ratio and micro-element content of the fossil sediments can be regarded as the same in the material of different boreholes, moreover, the concentrations of micro-elements in the fossil material exceeds that of the recent sediments by several magnitudes.
- Grain size pattern is in close correlation with the changes of the measured micro-element concentrations.
- The concentration of the majority of the measured elements did not exceed either the limits of the law or the limits based on the geo-accumulation index. Concentrations of silver and cadmium higher than the limit can be explained by the geological conditions of the source region of the river and also by the very low limits set by the legislation (Ag: 2 mg/kg, Cd: 1 mg/kg).
- Reworking of the anthropogenic load trapped in various sediment bodies of the bed can be assumed resulting probably in the wash-out of those parts of the mid-channel bars that accumulate anthropogenic load and thus in the mixing of sediments and the ultimate dilution of the anthropogenic load.

Although the presence of anthropogenic load in the sediments of the Kismarja section of Berettyó

River cannot be excluded based on the studied grain size distributions and micro-element concentrations and the above reasons, it can be regarded as subordinate due to the dominance of elements originated from geological sources.

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