An assessment of in-channel alluvia (aits) in the Suceava River near Suceava city, NE Romania

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ABSTRACT: A study was conducted to investigate the change in physical and geochemical composition of alluvial sediments in three selected aits along Suceava River, upstream and downstream of the Suceava urban area. Water chemical analyses were also carried out. Pit profiles were dug for each ait to assess the variability of sediment composition with depth. We used grain size distribution, organic matter and carbonate content, elemental geochemistry and magnetic properties to highlight vertical and horizontal variability of the selected parameters. Results show spatially distinct sources for the alluvial material deposited as aits, i.e., input of finer sediments in the upstream part (main river) and coarser material deposited in the downstream part (tributaries). Sediments and water downstream of Cetății Creek are characterized by high organic content, sulphur and heavy metal concentrations, resulting primarily from the discharge of the polluted waters of this tributary into Suceava River.

KEY WORDS: alluvial sediments, magnetic properties, geochemistry, particle-size, pollution.

1. Introduction

Alluvial deposits are a direct result of river activity and an indirect result of various environmental factors such as the climatic regime, landscape morphology, vegetation and land-use (Gurnell et al., 2001). River sediments are a combination of eroded material from catchment rocks and soils displaced and carried both by the main river and its natural and artificial tributaries. This material is separated during transport mainly according to its grain-size and shape, each size fraction having a distinct transportation pathway along the channel (Livarciuc and Rădoane, 2017). The grain size of riverbed alluvia can thus carry important information about flow rates, erosion and steady states (Dade and Friend, 1998). In addition, geochemical and mineral magnetic properties of sediments deposited in a river channel and floodplain, along with their organic matter and carbonate content, have the potential to indicate sediment sources and assess the degree of anthropogenic pollution (Swennen and Sluys, 2002; Hürkamp et al., 2009; Famera et al., 2018). Such issues are important when establishing sediment displacement and mobilisation, the persistence of pollutants and their biological accumulation (Salomons and Brils, 2004).

Determination of magnetic susceptibility and remanence in sediment samples is a rapid and non-destructive method (Thompson and Oldfield, 1986). It therefore allows a multitude of sampling sites to be analysed in order to efficiently discriminate polluted areas where additional, more expensive analysis might be undertaken. Geochemical assessment via X-ray fluorescence spectroscopy (XRF) is likewise fast and non-destructive (Boyle, 2000); it provides estimates of the elemental composition of sediments permitting the efficient screening and estimation of anthropogenic vs. lithogenic contributions to river sediments.

Although the use of these proxies on river sediments for environmental quality assessment has a long history in Western Europe, to date such studies are scarce in Romania and mainly restricted to areas affected by mining industries. For example, magnetic susceptibility screening was employed in the Baia Mare area as a pilot study to assess the degree of river pollution after tailing dam failure in 2000 (Wehland et al., 2002). Contamination of river sediments with heavy metals resulted from mining was determined via XRF along the Bistrita River and its tributaries (Maftei et al., 2014; 2018; Petrescu et al., 2010) and also in protected areas such as the Danube Delta (Vignati et al., 2013).

A combined use of these proxies can therefore provide a comprehensive picture of sediment sources, dynamics within the channel and sediment characteristics. To date, no such studies have been undertaken on the alluvial sediments of Suceava River (north-eastern Romania) for environmental assessment purposes, although this river crosses important urban areas and is used for drinking water, irrigation, fishing, flood control and industries.

This study aims to assess the variability of physical and geochemical properties in vertical profiles of recent alluvia deposited as aits along the Suceava River with the urban area of the city of Suceava. To our knowledge, there is no study that covers this topic for our study area. This hampers potential assessments of the anthropogenic influence related to urban settlements on river behaviour and sediment quality in the Suceava area.

2. Study area

The study area is located in north-eastern Romania and comprises a sector of the Suceava River channel, upstream to downstream of the city. Within the city, the Suceava River has an average flow rate of 16.9 m^3/s at Ițcani gauge (Briciu, 2017). The riverbed in the urban area is composed mainly of gravel, sands, clays and occasionally sandstone strata, reflecting the dominant lithology of the catchment - conglomerates, shales, sandstones, marls, clays and sands (Oprea, 2014). The lithology of the study area has a complex distribution of rocks due to fluvial terraces and alluvial fans (Martiniuc and Băcăuanu, 1960). Where flow conditions and sediment input from tributaries have favoured sediment accumulation, a number of aits have formed in the main river channel. Three of these aits (sand and gravel bars with herbaceous and shrub vegetation) were selected along the river in October 2012 for the analysis of alluvial deposits. They are located upstream and immediately downstream of Suceava city area, at the mouth of two natural and one artificial tributaries. The uppermost ait (H) was formed at Hătnuța River mouth (47°41'34"N, 26°9'54"E), the middle ait (C) in front of Cetății Creek mouth (47°39'9"N, 26°16'42"E), and the lowermost ait (W) is found at the point where Suceava city wastewater treatment plant discharges into the Suceava River (47°39'3"N, 26°17'16"E) (Fig. 1). The aits are linked to the surrounding floodplain through a basal layer dominated by boulders and pebbles. Above this layer there lie strata with a higher concentration of finer constituents, e.g. sands, clays (Fig. 2).

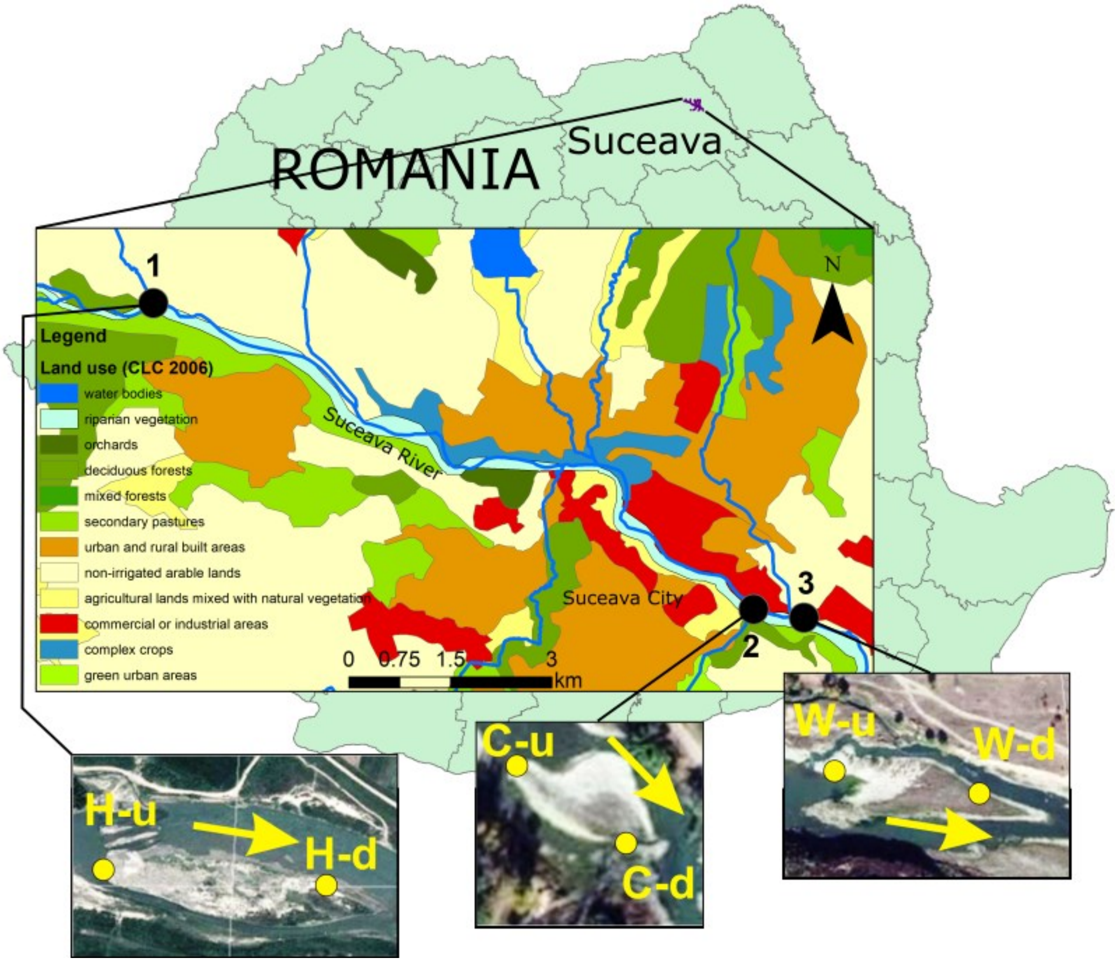

Figure 1 Location of the studied aits (representative mosaic of land uses for the entire Suceava Plateau) and details of the three selected aits (u – upstream; d – downstream), as they appeared in 2012 (details concerning the size of each ait are presented in the text below); the arrows indicate the direction of flow (Satellite imagery source: Google, DigitalGlobe 2011-2012; Land use data source: European Environment Agency (2006). This figure is available in colour online at www.georeview.ro.

Some morphometric details of the aits in 2012 were as follows: uppermost ait – length (L)=408 m, width (W)=98 m; middle ait - L=103 m, W=35 m; lowermost ait - L=333 m, W=61 m. The maximum height of each ait at the time of sampling did not exceed 0.5 m above the base flow water level of Suceava River.

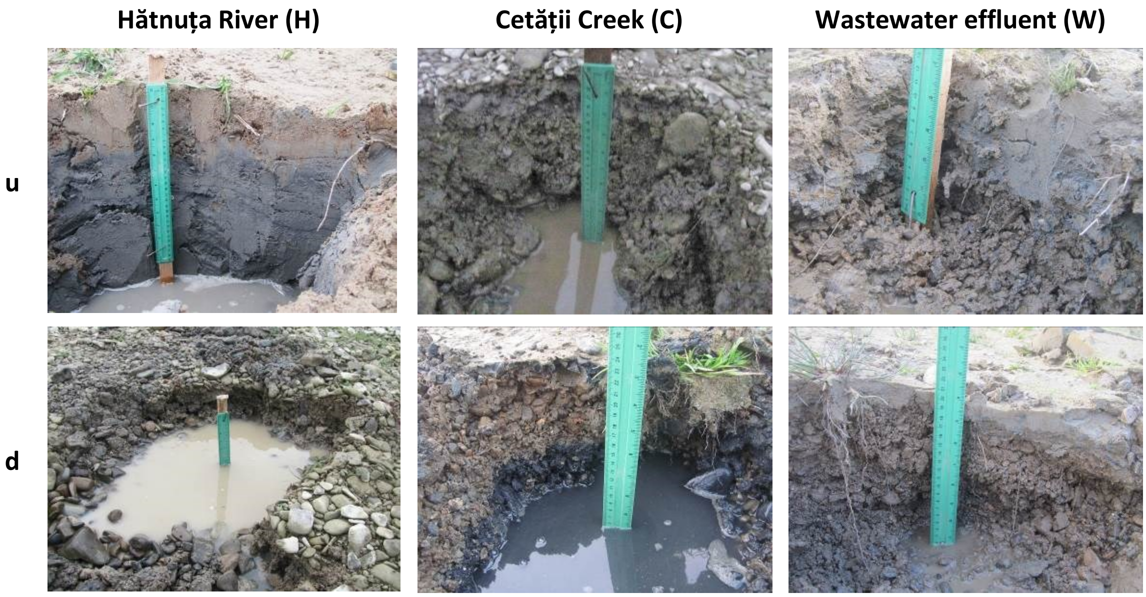

Figure 2. Details of the pits dug in the three selected aits (H, C, W): u - upstream sampling points, d - downstream sampling points. This figure is available in colour online at www.georeview.ro

3. Methods and data

The sediment samples were extracted at the end of October 2012 in polyethylene jars. Two pit profiles were dug on each ait, at the upstream and downstream extremities. The depth interval of sampling followed the changes in visual properties of sediment layers in each pit. Sediment samples were dried at 37˚C for two days and sieved, in order to permit further analyses only on particles < 1 mm diameter (the finest sediment fraction). The proportion of particles >1mm in each sample was determined, and this size fraction, comprising coarse sand, granules and pebbles (Wentworth, 1922), is referred to in this paper as ”gravel“. Samples were weighed before and after drying, in order to determine their water content (humidity).

Organic matter and carbonate content were assessed via a loss-on-ignition method after the procedure described by Heiri et al. (2001). For organic matter determination, sediment samples were dried for 24 h at 105°C and combusted for 4 h at 550°C, whereas estimation of carbonate content was performed by further burning the remaining ashes at 925°C for 2 hours. The weight lost after each stage of sediment burning was expressed as a percentage of the initial dry weight. Particle size was automatically measured on the ashed samples resulting from carbonate content determination via a Horiba Laser Scattering Particle Size Analyzer (Partica LA-950).

Sediment elemental geochemistry was determined with a non-destructive Niton XL3t 900 X-Ray Fluorescence analyser (fpXRF) and follows the method presented in Hutchinson et al. (2016). As a Certified Reference Material (CRM) we used NCS DC73308. For mineral magnetic characterisation, a Bartington Instruments Ltd MS2 susceptibility meter with B sensor was employed to determine magnetic susceptibility. Low frequency mass specific (χ) and frequency dependent susceptibility ($\chi FD\%$) were obtained (Dearing, 1999). To determine magnetic remanence, a Molspin AF Demagnetiser was used for Anhysteretic Remanent Magnetisation (ARM). The Saturated

Isothermal Remanent Magnetisation (SIRM) (magnetic field 1.0 T) and Isothermal Remanent Magnetisation (IRM) backfields (-20mT, -40mT, -100mT and -300mT) were conveyed with a Molspin Ltd Pulse Magnetiser. For ARM, SIRM and backfields, the resulting magnetic remanence was measured with a Minispin Fluxgate Magnetometer (Hutchinson et al., 2016) and mass normalised. Soft and hard (HIRM) magnetic components were calculated following Thompson and Oldfield (1986).

Water samples were taken from Suceava River near the sediment sampling points during the same day and analysed in the laboratory within 24 hours of sampling by using a Hach-Lange DR2800 spectrophotometer. The analysed chemical water parameters (mg/L) are: orthophosphates (PO_4^{3-}), nitrogen from nitrates and nitrites (NO_3^- - N, NO_2^- - N), sulfates (SO_4^{2-}) and sulfides (S^{2-}). Reagents and methods are identical to those described in Briciu et al. (2016).

4. Results and discussion

We observed an inverse relationship between the humidity and gravel percentages in our sediment samples (Fig. 3). When comparing the average values of the upstream and downstream samples, no relevant difference between the upper and the lower part of the aits is apparent: gravel - 45.6% in the upstream samples and 46.6% in the downstream samples; humidity - 12.8% and 12.1%, respectively. 10 of the analysed samples show average gravel percentages around 72.78%, whereas the remaining 6 samples have gravel percentages around 1.83%. Thus, high percentages of gravel correspond to lower humidity percentages and vice versa. The statistical distribution of the humidity and gravel percentages is thus strongly bimodal, probably suggesting self-sustaining sedimentary processes during the deposition of the alluvial material.

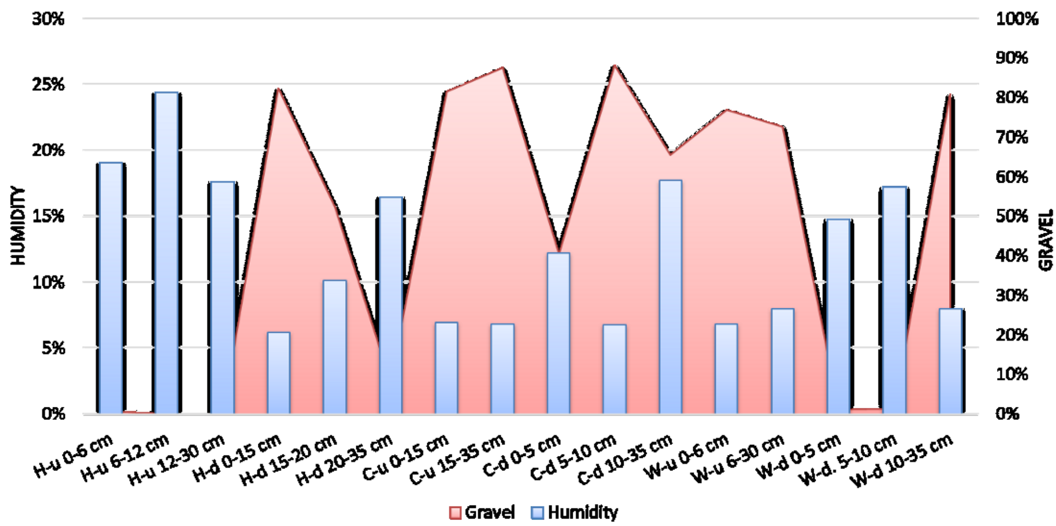

Figure 3 Humidity and gravel percentage variation in the selected sites and depths. This figure is available in colour online at www.georeview.ro.

Each ait formed at the mouth of tributaries, immediately downstream of rapids imposed by the coarser material in the riverbed. We therefore expected sediment composition of these aits to be a reflection of the input from both Suceava river and tributaries. Furthermore, since the wastewater treatment plant effluent (W) does not provide any significant alluvial input into the Suceava river, we expected that the development of the lowermost ait is connected to sediment input from Suceava River and Cetății Creek. Indeed, within each island, the particle size distribution of sediment grains <1 mm tends to have 2 types of shapes: a bimodal one in the upstream part of the islands and a generally unimodal one in the downstream part (Fig. 4). The two peaks of the bimodal distribution correspond to particle size diameters ranging from 451 to 678 µm for one peak (592 µm as mode) and from 34 to 101 µm for the other peak (58 and 101 µm as modes). The unimodal distribution peaks range from 133 to 517 µm (133 µm as mode). The most probable cause of the change from bimodality to unimodality is the solid discharge into Suceava River from its nearby tributaries: Hătnuța River and Cetății Creek. The higher size particles are probably originated mainly from the tributaries' catchments during high waters, whilst the finer particles are provided mainly by Suceava River and deposited, as expected, in the upstream part of the islands, behind the coarse material carried by the tributaries. This idea is confirmed by the analyses carried out by Briciu (2017) on the particle size distribution of Dragomirna Lake, a reservoir within Suceava city administrative area. This lake has sediments deposited by the input waters from Dragomirna River (direct discharge) and Suceava River (pumped water). The upper half column of sediments shows a weaker bimodal distribution compared to the lower half, when the water input from Suceava River was much more significant.

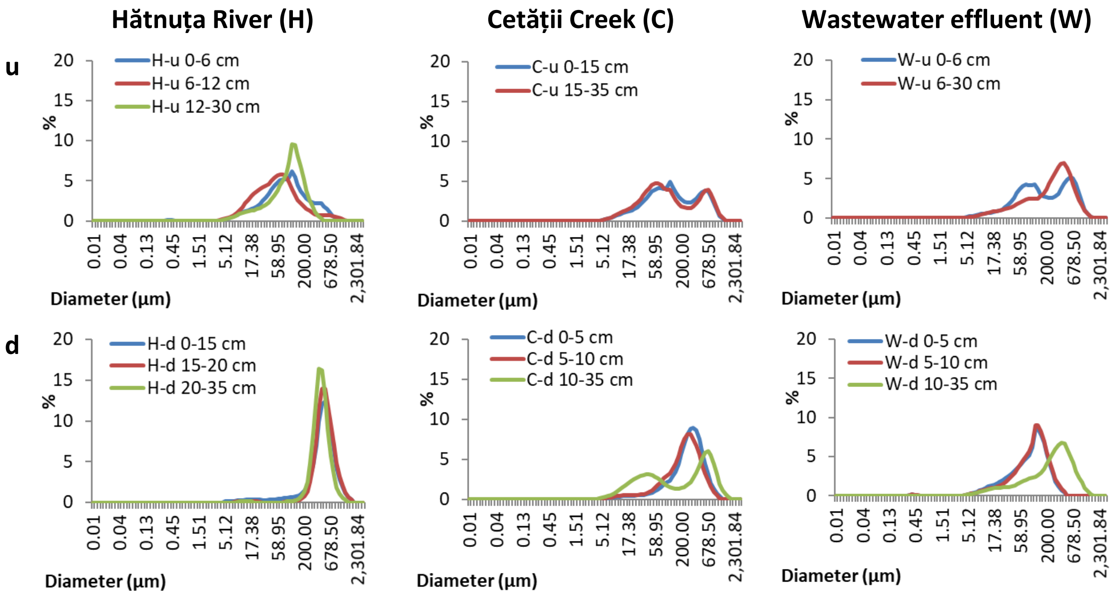

Figure 4 Particle size distribution of sediment grains <1 mm at the three selected aits (H, C, W): u - upstream sampling points, d - downstream sampling points. This figure is available in colour online at www.georeview.ro.

Median grain size distribution (D50) also confirms the existence of a clear demarcation in terms of sediment sources between the upstream and the downstream parts of the islands; D90 (intercept for 90% distribution of grain diameter) indicates the clear separate evolution of H-u, a similar distribution being found only in the upper layer of W-d (Fig. 5).

The structure and evolution of these aits are also interconnected with the activity of the gravel pits along Suceava River. Gravel and sand extraction from riverbeds was already shown as environmentally harmful at both channel and catchment level, especially when associated with other human activities that reduce upstream sediment delivery (Rinaldi et al., 2005). There are two important and almost permanently active gravel pits on Suceava River upstream and downstream of the uppermost ait. Their activity, together with the dam at Mihoveni (between the upper and middle aits), have likely influenced the alluvial dynamics along the river in the study area. These factors have likely induced supplementary river erosion and local accumulation of finer sediment, which is also highlighted by the particle size distribution for the upstream part of the aits (Fig. 5). The middle and lowermost aits emerged mainly during the high waters of 2008 and 2010, whilst the uppermost ait appears much older. The importance of tributaries as sediment contributors is also highlighted by: i) the relative temporal stability of the upstream ait, ii) the fact that the middle ait is currently a part of the alluvial fan of Cetății Creek (which provides important sediment supply), and iii) enhanced water erosion at the lowermost ait (at the mouth of the wastewater effluent with minimum alluvial input) where half of the ait has already been washed away.

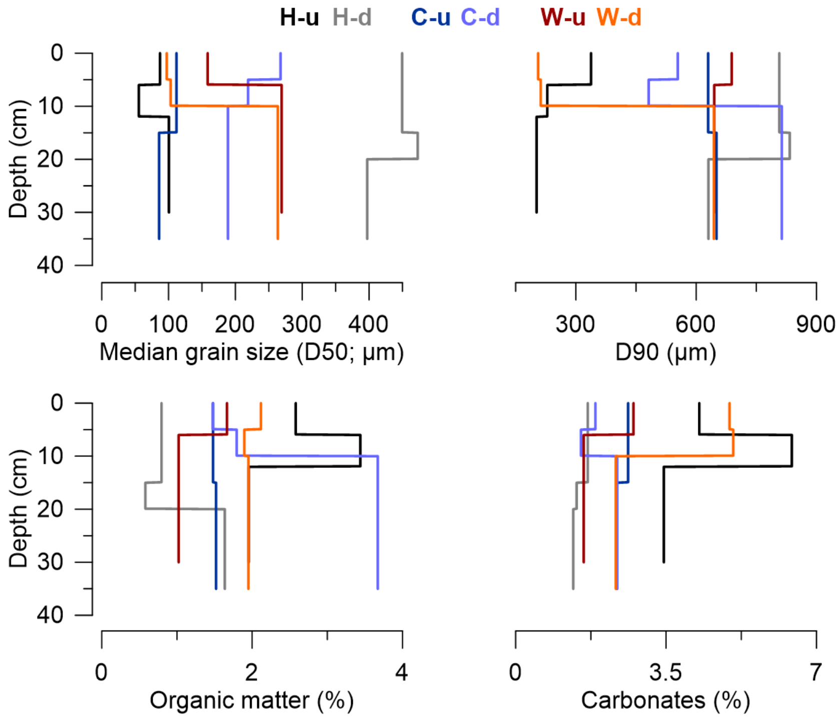

Figure 5 Grain size variability, organic matter and carbonate content in the selected sites and depths. This figure is available in colour online at www.georeview.ro.

Results show very small percentages for organic matter content in sediments, at all sampling points and depths (<4%) (Fig. 5). Among these, the highest values characterize deeper sediments of C-d profile, where an anoxic bottom layer with decomposing organic matter can be observed (Fig. 2). The excess organic content is likely caused by the polluted waters discharged by Cetății Creek, which at that time (prior to 2012) received untreated wastewater from Suceava city (not all wastewaters were collected by the wastewater treatment plant to be then discharged into Suceava River). In support of this hypothesis, pollution of Suceava River at the Cetății Creek discharge point can be easily observed in water composition which reveals high concentrations of orthophosphates, nitrites and sulfides (Fig. 6); direct (visual) evidence was also available at the Cetății Creek sampling point (Fig. 7). There is a positive correlation between the sulfide and sulfate concentrations, suggesting that sulfates are mostly a result of the sulfides oxidisation. This rule applies especially when high sulfides concentrations are present (downstream of the wastewater treatment plant and Cetății Creek).

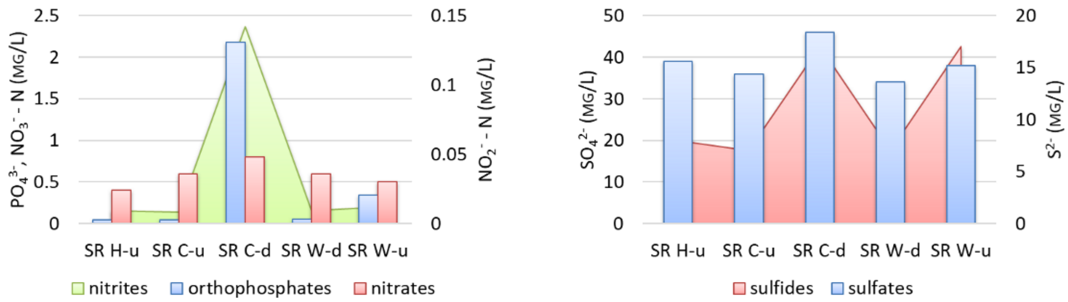

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Figure 6 Values of chemical parameters measured in water samples from Suceava River (SR) near some sediment sampling sites. This figure is available in colour online at www.georeview.ro.

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Figure 7 Examples of the anoxic, sulfide-rich layer in Suceava River streambed, downstream of Cetății Creek discharge point. The anoxic layer is found under a thin layer of fresh alluvial deposits. This figure is available in colour online at www.georeview.ro.

The carbonates content shows high values in the upper layers of W-d and H-u, in opposition with the quasi-linear, low values at the other measuring points (Fig. 5). There is an almost identical evolution between carbonates content and Ca concentrations at W-d and H-u indicating a $CaCO_3$-rich alluvium (Fig. 8) This alluvium is characteristic of samples with smaller grain-size, thus likely deposited by the Suceava River (Fig. 4). Rădoane et al. (2001) showed that, within Suceava city, the Suceava River streambed is mainly composed of silicate rich- and lime rich sandstones. The latter are probably the sources of the finer alluvial material rich in $CaCO_3$. Sr presents the same distribution as Ca, suggesting that there are supplementary carbonate particles, probably of carbonate strontianite ($SrCO_3$) type.

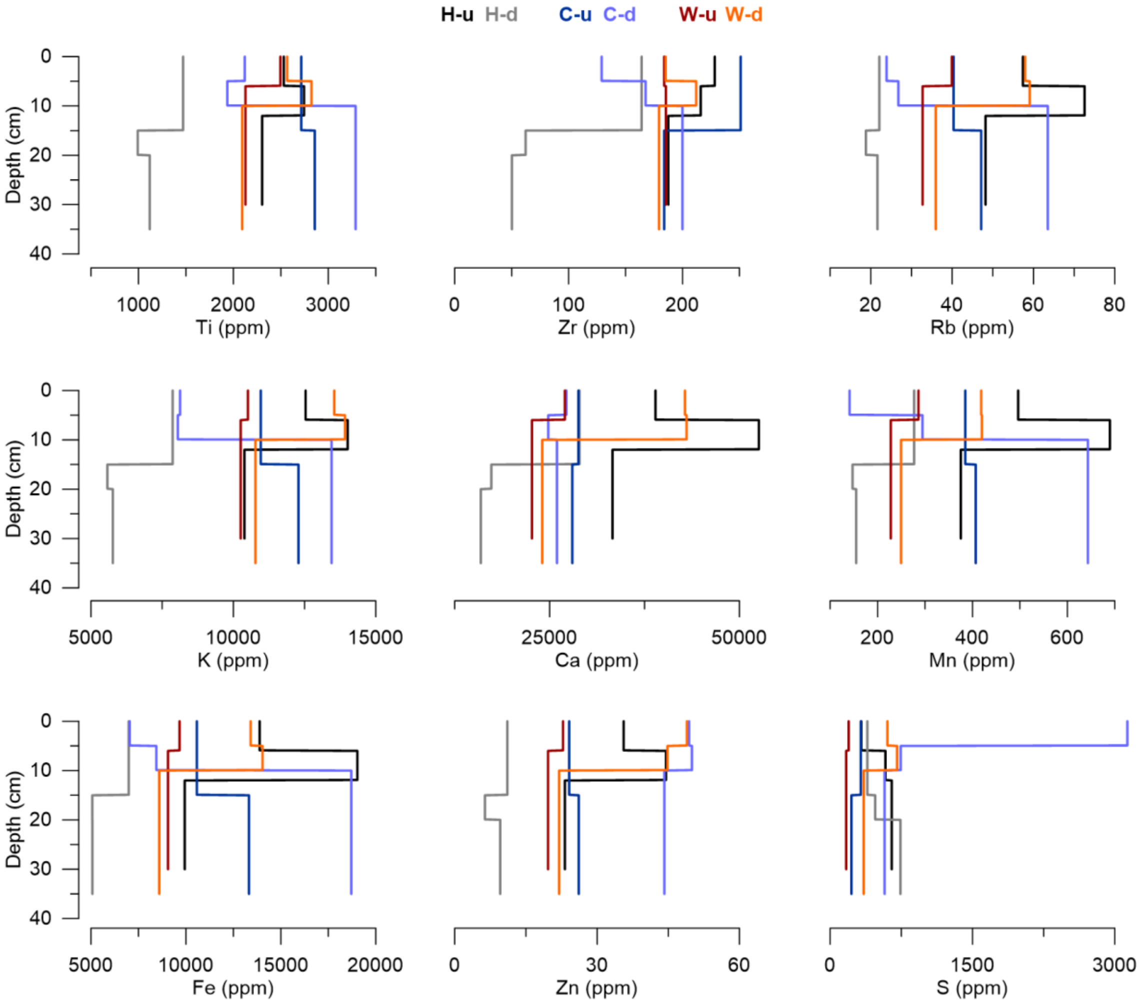

Figure 8 Results of the XRF chemical analysis. This figure is available in colour online at www.georeview.ro.

In terms of geochemical composition, the concentration of individual elements strongly varies with sampling points and depths (Fig. 8). Most major and trace elements are negatively corelated with grain-size, and the correlation is strongest for D50 (Table 1). Major (e.g. K, Ti, Ca, Fe, Rb) and trace elements (Cu, Zn) are positively correlated among themselves, which would indicate a common mechanism regulating their abundance, associated with the finest sediment fraction.

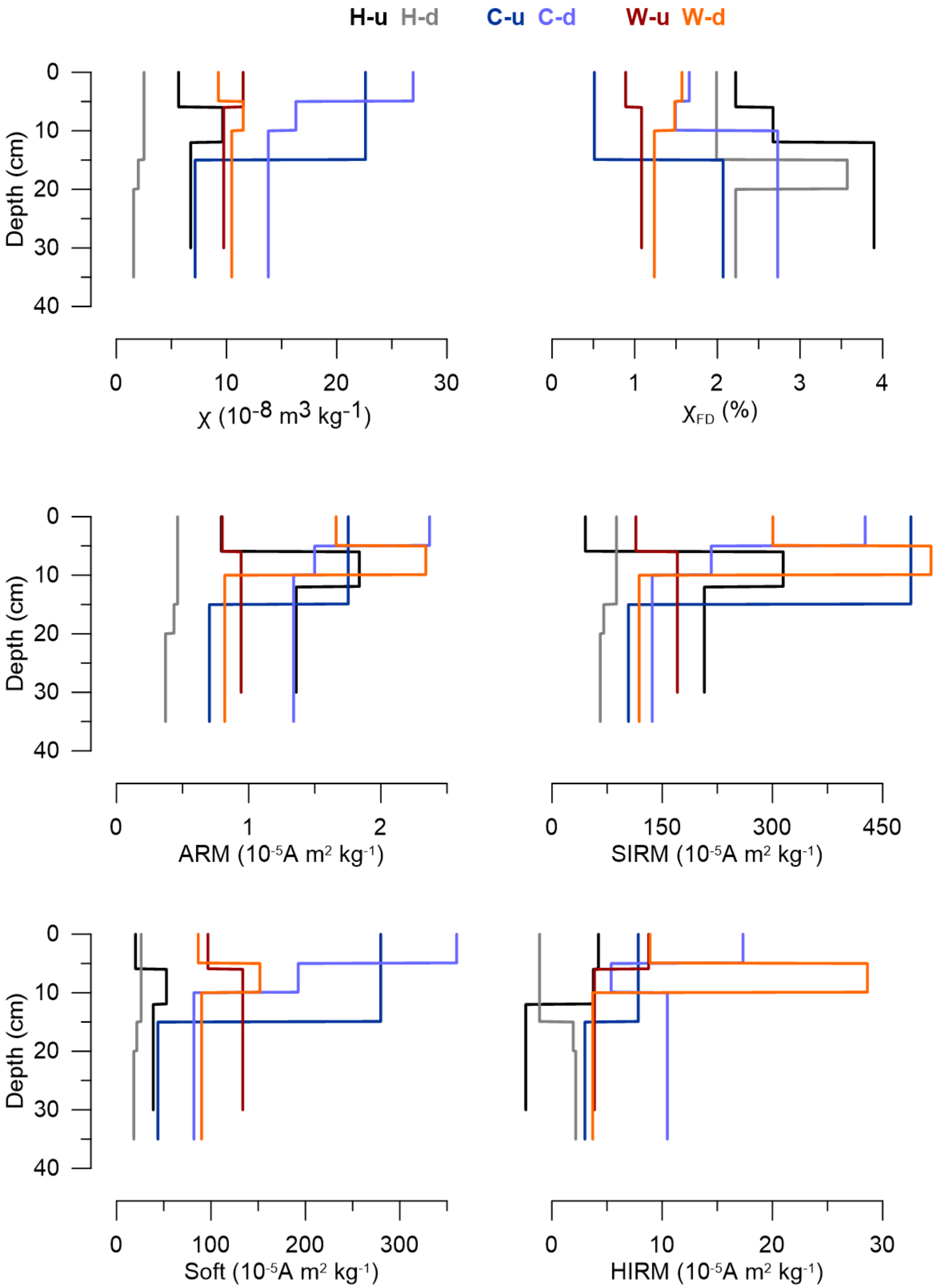

Figure 9. Magnetic mineral properties of the alluvial samples. This figure is available in colour online at www.georeview.ro

Altogether, this suggests that sedimentation of finer particles in the study area is highly controlled by clay mineral abundance (e.g. Dala et al., 2004). Mn shows significant positive correlation with Fe (r=0.97), which suggests the occurrence of Mn-Fe components in oxide phase. A positive correlation of trace metals (Cu, Zn) with Fe and Mn, but also with organic matter content suggests that Fe-Mn oxides and organic matter play an important role in regulating metal concentrations in the river channel. As it can be observed in Fig. 9, there is an almost direct proportionality between some elements in all sampling points. There is a Fe-Mn-Rb-Ti-K pair and a Ca-Sr pair, suggesting that these elements probably form minerals/pairs.

Mineral magnetic properties of the alluvial samples are similarly highly variable (Fig. 9). Frequency dependent susceptibility (χFD) values close to 4% for H-d (15-20), H-u (12-30, 6-12) and C-d (10-35) point to a low concentration of superparamagnetic (SPM) magnetite grains formed during pedogenesis (Dearing, 1999), thus indicating top soil erosion from Suceava River catchment. The content of the SPM grains in the upper sediment layers is considerably reduced, probably due to outwash or downward eluviation within the sediment profile.

Mass susceptibility (χ) values indicate a higher input of magnetic minerals in the upper layers of C-u, C-d and W-u, W-d, which are also magnetically soft (Fig. 9). χ appears inversely correlated with grain size at C-u and W-u (magnetic grains occur in the clay fraction), whereas for C-d, χ is directly influenced by grain size (magnetic grains occur in the silt and fine sand fractions).

SIRM is highest for C-u and C-d (>400 x $10^{-5}Am^2kg^{-1}$), where the proportion of magnetically soft minerals is maximum (Fig. 7), but also for the top part of W-d, where magnetically hard minerals dominate the remanence. In the Cetății Creek and wastewater effluent areas (C-d, W-d), elevated concentrations of Zn, Cu and S are also observed (Fig. 8; Table 1). Magnetic parameters show a statistically significant positive correlation with these elements (Table 1). Furthermore, the proportion of magnetically soft minerals and the variability of ARM appear positively correlated with S concentration, but also to Ca and Sr concentration (Table 1).

Altogether, this shows an increased concentration of ferrimagnetic minerals of anthropogenic origin and some heavy metals in the Cetății Creek area, likely due to the enhancement of iron oxides and sulfides derived from water and air pollution (emissions from the thermal power plant, discharge of untreated wastewaters and pluvial drainage from surface runoff).

5. Conclusions

Our analyses indicate a strong variability of sediment properties within and between the six sampling sites. Sediment deposition patterns in the aits of the Suceava river channel appear to be influenced by both the main river (input of finer sediments in the upstream part of the aits) and tributaries (coarser material deposited in the downstream part), and this sediment dynamics is likely influenced by the presence of gravel pits and dams on the river course. The anthropogenic influence on the sediment composition is evident downstream of Cetății Creek mouth, where high organic and sulfide concentrations, along with elevated concentrations for Zn and Cu were measured, as result of water pollution. Further work is necessary to understand the structure and composition of alluvial sediments in the studied aits, such as the study of sediment samples from Suceava riverbed in places where the sedimentary influence of local tributaries is absent or weak.

Table 1 Correlation matrix of selected parameters.

S	K	Ca	Ti	Mn	Fe	Cu	Zn	Rb	Sr	Zr	D90	D10	D50	CC%	OM %	SOFT	-300 mT	-100 mT	-40 mT	-20 mT	SIRM	ARM	χ FD	
0.59	0.17	0.07	0.43	0.02	0.12	0.44	0.59	0.04	0.01	0.36	-0.05	-0.48	-0.32	-0.02	0.14	0.95	0.70	0.76	0.35	0.14	0.71	0.75	-0.50	**X**
0.03	-0.15	0.04	-0.21	0.18	0.08	0.09	-0.16	0.12	0.09	-0.35	-0.11	0.35	0.15	0.09	0.19	-0.56	-0.35	-0.39	0.03	0.00	-0.35	-0.20	1	**χ FD**
0.56	0.47	0.57	0.54	0.32	0.41	0.52	0.80	0.42	0.46	0.44	-0.58	-0.54	-0.54	0.51	0.35	0.71	0.91	0.92	0.82	0.68	0.92	1		**ARM**
0.43	0.35	0.47	0.41	0.16	0.23	0.26	0.55	0.27	0.34	0.40	-0.47	-0.39	-0.43	0.42	0.09	0.74	1.00	1.00	0.73	0.77	1			**SIRM**
0.02	0.55	0.73	0.41	0.45	0.47	0.21	0.38	0.57	0.66	0.41	-0.68	-0.31	-0.51	0.75	0.26	0.15	0.77	0.71	0.72	1				**-20 mT**
0.49	0.32	0.63	0.25	0.20	0.28	0.26	0.62	0.34	0.47	0.16	-0.70	-0.30	-0.37	0.58	0.22	0.38	0.73	0.73	1					**-40 mT**
0.49	0.30	0.42	0.38	0.11	0.18	0.26	0.57	0.21	0.28	0.38	-0.44	-0.39	-0.40	0.36	0.06	0.80	0.99	1						**-100 mT**
0.42	0.34	0.47	0.40	0.16	0.23	0.26	0.54	0.26	0.34	0.40	-0.47	-0.39	-0.43	0.42	0.09	0.74	1							**-300 mT**
0.65	-0.03	-0.05	0.20	-0.22	-0.13	0.19	0.46	-0.19	-0.16	0.19	-0.02	-0.28	-0.13	-0.15	-0.13	1								**SOFT**
-0.03	0.69	0.56	0.68	0.85	0.85	0.81	0.62	0.82	0.67	0.47	-0.40	-0.54	-0.61	0.61	1									**OM%**
-0.11	0.78	0.96	0.57	0.73	0.74	0.47	0.53	0.86	0.95	0.57	-0.81	-0.53	-0.73	1										**CC%**
0.12	-0.85	-0.72	-0.87	-0.71	-0.76	-0.60	-0.62	-0.83	-0.81	-0.79	0.68	0.87	1											**D50**
0.07	-0.84	-0.59	-0.89	-0.61	-0.69	-0.68	-0.64	-0.70	-0.66	-0.89	0.48	1												**D10**
-0.10	-0.53	-0.80	-0.36	-0.41	-0.42	-0.25	-0.57	-0.60	-0.71	-0.40	1													**D90**
-0.29	0.82	0.64	0.83	0.70	0.70	0.62	0.46	0.70	0.71	1														**Zr**
-0.20	0.88	0.95	0.72	0.86	0.87	0.63	0.55	0.93	1															**Sr**
-0.24	0.94	0.81	0.84	0.93	0.96	0.72	0.57	1																**Rb**
0.43	0.59	0.60	0.63	0.52	0.61	0.70	1																	**Zn**
0.20	0.69	0.55	0.79	0.80	0.82	1																		**Cu**
-0.21	0.91	0.73	0.87	0.97	1																			**Fe**
-0.29	0.84	0.73	0.78	1																				**Mn**
-0.10	0.92	0.59	1																					**Ti**
-0.02	0.77	1																						**Ca**
-0.24	1																							**K**

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