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ASSESSMENT OF LANDSCAPE SENSITIVITY BASED ON GEOCHEMICAL CHARACTERISTICS OF SEDIMENTS (KREMNA BASIN, SERBIA)

Kašanin-Grubin Milica*, Šajnović Aleksandra*, Perunović Tamara, Stojanović Ksenija**, Simić Vladimir***, Gajica Gordana*, Brčeski Ilija****

*University of Belgrade, Institute of Chemistry, Technology and Metallurgy IChTM, Njegoševa 12, 11000 Belgrade, Serbia, e-mail: mkasaningrubin@chem.bg.ac.rs, sajnovica@chem.bg.ac.rs, ggajica@chem.bg.ac.rs
**University of Belgrade, Faculty of Chemistry, Studentski trg 12-16, 11000 Belgrade, Serbia, e-mail: tamara.perunovic@gmail.com, ksenija@chem.bg.ac.rs, ibrceski@chem.bg.ac.rs
***University of Belgrade, Faculty of Mining and Geology, Djušina 7, 11000 Belgrade, Serbia, e-mail: simicv@rgf.bg.ac.rs

Abstract

The scale of human impacts on the natural environment is now considerably larger than at any point in history. The concept of geomorphic sensitivity can help to understand the rate, magnitude and nature of landscape adjustment to perturbation in a given natural system. Aim of this research is to show that geochemical and mineralogical data are important factors in determining landscape sensitivity. To test the suggested premise Neogene lacustrine Kremna basin (Serbia) was selected since sediments found in lacustrine basins are usually prone to dispersion and erosion. Furthermore, lacustrine basins often bear fossil and mineral resources and because of that are often undergoing land use changes. For the purpose of this study, samples of serpentinite, carbonates, marly carbonates, oil shale and tuff were analyzed. Besides mineralogical and petrographic analyses, samples were subjected to the weathering experiments. Obtained results indicate that marly carbonates and tuff are most prone to dispersion primarily due to presence of clay minerals. However, oil shale which also contains clay minerals showed minor leaching characteristics due to high content of organic matter. It can be concluded that mineralogical and geochemical characteristics are important for determining landscape sensitivity to erosion processes of an area.

Key words: landscape sensitivity, lacustrine sediments, mineralogy, geochemistry, leaching

INTRODUCTION

Human impact on their surrounding natural environment started thousands of years ago, however, these impacts for a long time were short-lived changes, localized in time and space. The scale of human impacts on the natural environment is now considerably larger than at any point in history and this might mean that human activities and landscape processes should not be treated separately but as a coupled system (Werner, McNamara, 2007). Landscapes respond to changes in external drivers in a complex and indirect way and the effects can last up to thousands, or even millions of years (Schoorl et al., 2014).

The concept of geomorphic sensitivity was introduced by Brunsden and Thornes (1979) as an aid to understand how natural systems respond to environmental change. This concept generally differentiates robust and sensitive landscapes. However, it is not necessary that the whole system is sensitive to an imposed change, but one or more components of the landscape can be sensitive in certain circumstances, and change in that component can trigger instability in other parts of the system. Furthermore, this makes the system conditionally unstable when a quick and irreversible change, caused by perturbations in the controlling environmental processes, might occur (Thomas, 2001).

Geomorphic response to natural of anthropogenically imposed disturbance shows evident differences in the rate, magnitude and nature of landscape adjustment to perturbation (Miller et al., 2011). Concept of sensitivity can help in understanding these differences, i.e. (1) the likelihood that an area will respond to an imposed disturbance, (2) the time, duration, rate and nature of the response and (3) the potential for a given system to be stabilized (Downs, Gregory, 2004).

So far landscape sensitivity was studied from different points of interest. Landscape sensitivity in general was of main interest in a number of studies (e.g. Brunsden, Thornes, 1979; Thomas, 2001; Usher, 2001) but other, more specific, aspects were also considered. For example, Miles et al. (2001) discussed the ecological view of this concept, Harvey (2001) assessed the role of hillslope/channel coupling for determining the system sensitivity, Werritty and Leys (2001) and Florsheim et al. (2013) analyzed sensitivity of fluvial systems and Grieve (2001) sensitivity of soil properties. A rare attempt to quantify the concept of landscape sensitivity was done by Kereny and Csorba (1991). Based on the climatic sensitivity of four landscape factors (the water budget of soil, thickness of soil, insolation and land use), these authors suggested five sensitivity categories: no sensitivity to climate variability, moderate sensitivity, average sensitivity, high sensitivity and very high sensitivity. Hua et al. (2015) also studied influence of land use and land cover change from grassland to forest in China on climate regimes and showed that there are evident changes in temperature and precipitation which are more evident during summer and fall.

So far none of the studies took into consideration the geological setting as a factor of landscape sensitivity. Geology, landscape and humans have very important interactions and geological data provide essential scientific information for society and economy. Understanding the role that parent material plays in different processes within soil-mantled landscapes under variable soil production and vegetation dynamics is still limited (Booth, Brayson, 2011). Geological setting, primarily type of rock that is the base for the geomorphic processes can be considered as a constant in a given landscape.

Rock characteristics determine the soil and sediment unique geochemical composition and if this composition does not change then the spatial origin of transported material may be determined (Laceby et al., 2015).

On the other hand, soil formation and vegetation cover develop and change depending on weathering and erosion processes as well as changing climate. Since “the autogenic factor inherent in a natural systems add to the difficulty in defining a single cause of geomorphic change” (Macklin et al., 2012), it makes the study of geological setting regarding landscape sensitivity even more important.

Aim of this research is to show that geochemical and mineralogical data are important factors in determining landscape sensitivity. Once exposed to surface conditions rocks undergo different weathering processes. The dominant weathering process depends on climate conditions and rock composition and structure. Physical weathering will cause breakage of material to shards and depending on the slope steepness, runoff will move the material downslope. On the other hand, chemical and biological weathering is important for potential migration of major and trace elements and their transfer to water and/or soil. Sorption at the solid/solution interface is the prevailing physico-chemical process which controls the ability of soils to adsorb and retain heavy metals. Weathering and pedogenetic processes will also determine soil adsorption capacity of heavy metals, not only at the scale of the bulk sample, but also at the specific weathering microsystems including the rock-forming minerals (Caillaud et al., 2006).

To test the suggested premise that geochemical and mineralogical data are important factor for determining landscape sensitivity Neogene lacustrine Kremna basin (Serbia) was selected for three main reasons. First reason is that sediments found in lacustrine basins such as marls, limestones, dolomites with various quantities of clay minerals, tuffs and tuffaceous sediments, are usually prone to dispersion and erosion (e.g. Imeson, Verstraten, 1988; Canton et al., 2001; Faulkner et al., 2003; Kašanin-Grubin, Bryan, 2007). Second reason is the uniform geological setting since the whole basin lies on serpentinites. Third reason is the fact that lacustrine basins often bear fossil and mineral resources (coal, oil-shale and non-metallic minerals such as magnesite, borates and marls) and because of that are often undergoing land use changes. Taking all this into account, our hypothesis is that any type of land use change like deforestation or excavations would impose a great pressure on a sensitive landscape which would respond with notable geomorphic change.

MATERIAL AND METHOD

Study area

The Kremna basin, covering an area of 15 km^2, is a lacustrine basin of Zlatibor complex is located in southwest Serbia, about 200 km from Belgrade (Fig. 1). During the last few decades Kremna basin was studied for the occurrence of searlesite in the magnesite deposit (Živković, Stojanović, 1976), magnesite and dolomite (Obradović et al., 1994, 1995) and sepiolite and palygorskite clays (Kovačević, 1998). Zlatibor Mountain is one of the largest serpentinite massives in the Balkan Peninsula and is an ecologically exceptional area with 144 freshwater algae, 960 plant species, 120 medicinal herbs, 280 insect species, 10 amphibians and reptiles, 150 bird species and 54 mamal species (LBAP, 2011).

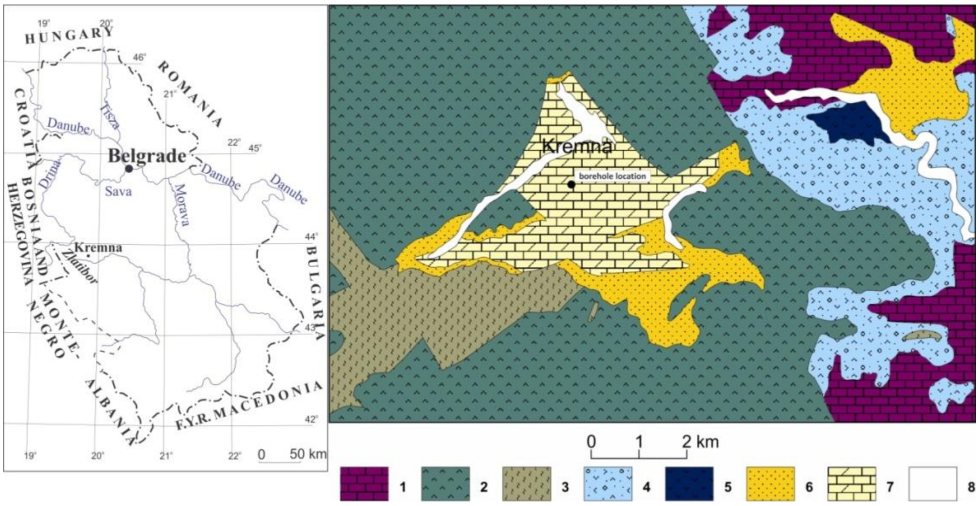

Fig. 1. Simplified geological map of the Kremna basin (modified after Basic Geologic Map of SFRJ, sheet Užice, 1:10000)

Legend: 1. Triassic carbonate rocks; 2. Hartzburgite; 3. Serpentinite; 4. Ophiolitic melange; 5. Diabase; 6. Lower Miocene alluvial sediments; 7. Lower Miocene lacustrine sediments; 8. Quaternary alluvial sediments (modified after Perunovic et al., 2014)

The landscape of the Kremna basin is hilly-mountainous with pastures, meadows and agriculture as dominant vegetation type. The area is sparsely populated with mountain villages which are dispersed and mostly isolated. Main water supply for villages is springs. Zlatibor Mountain, including Kremna, is characterized by a sub-mountainous climate. Snow is the main precipitation type from October until May, with an annual average of 110 days with snow-cover. Average annual temperature (1961-2012) is 7.5^{o}C, and average annual precipitation (1961-2012) is 988.3 mm (RHIS, 2014).

Analyzing the climatic data during the period 1961-2014 it is evident that since 1990 more profound change between colder and wetter, and warmer and drier years occur alternating every two to three years (Fig. 2). Yetemen et al. (2010) stressed out the critical role of climate on landscape processes in a sense that climatic fluctuations that can lead to replacing vegetation types could greatly intensify hydrological and geomorphic responses.

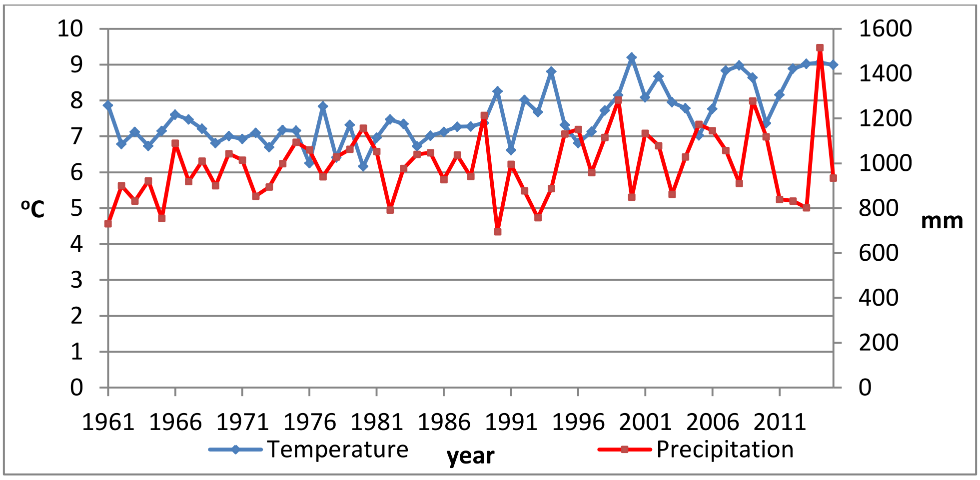

Fig. 2. Precipitation data for Zlatibor Mountain (1961-2015) (data obtained from the Republic Hydrometeorological Service of Serbia RHIS, 2016)

Geology of the Kremna basin is presented in Figure 1. The total thickness of sediments is about 350 m, and their age is Lower Miocene, Egenburgian-Otnangian (20.8-17 M years old) according to division for Paratethys (Rögl, 1996).

Perunović et al. (2014) studied geochemical characteristic of the Kremna basin sediments from one borehole (343 m depth) and distinguished upper and lower zone of the intrabasinal lacustrine facies. Dolomites and magnesites are dominant in the lower sequence (216-343 m), while in the upper facies sequence (13.5-216 m) carbonates containing both calcite and dolomite are prevalent (Perunović et al., 2014). Layers of volcanoclastic material are present throughout both sedimentary sequences.

In order to test whether the area of the Kremna basin is sensitive to possible anthropogenically imposed changes, mineralogical and geochemical characteristics of sediments were determined. Sediment sample selection was made based on the following criteria: representativeness in the sedimentary sequence and presence in the outcrops. Since the area of the Kremna basin is covered with vegetation, outcrops are rare and are mostly found at the basin edges. Beside serpentinite, using characteristic marker layers of oil shale and tuffs, 24 samples of the Kremna basin sediments were selected: 10 marls, 6 carbonates, 4 tuffs, 2 oil shale and 2 serpentinite samples. The reason for smaller number of samples of oil shale and serpentinite is that oil shale is present in only one layer in the sedimentary sequence and serpentinte is the underlying rock of the basin. To ensure that samples were not exposed to the natural weathering processes, specimens for the analyses and leaching experiment were taken from the borehole.

Methods

Prior to leaching experiment, mineralogical, petrographic and geochemical analyses were determined on all samples. Quarter of each sample was taken and homogenized and medium sample was separated for analyses. Concentrations of 10 elements (4 major and 6 trace elements) were analyzed using Inductively coupled plasma – optical emission spectroscopy (ICP-ES SPECTRO ARCOS instrument) and Inductively coupled plasma mass spectrometry (ICP-MS ELAN 9000 instrument). Accuracy and precision were ensured by using standard laboratory procedures and replicate measurements.

For determination of qualitative composition of the mineral part, X-ray generator PHILIPS Type PW 1729, and a diffractometer of the same manufacturer, type PW 1710 with genuine software processing (Philips APD) were used. Identification of minerals was performed on the basis of the reflection of the most frequent peaks and comparison with the database (Joint Committee on Powder Diffraction Standards, JCPDS-International Centre for Diffraction Data).

Scanning electron microscopy (SEM) analyses were performed on gold-coated thin sections. SEM was used for imaging and collecting chemical data, using a JEOL JSM–6610 LV Scanning Electron Microscope, equipped with an energy-dispersive spectrometer (EDS). Analyses were run at an accelerating voltage of 20 kV, and a working distance of 10 mm.

Content of organic carbon (Corg) was determined after the removal of carbonates from samples with diluted hydrochloric acid (1:3, v/v). The measurements were performed using a Vario EL III, CHNOS Elemental Analyser, Elementar Analysensysteme GmbH.

Electrical conductivity and pH were measured in solution made of 1 g of finely crushed sample and 10 ml of distilled water. Leaching experiment was conducted in controlled laboratory conditions in an apparatus specially designed for this purpose (Fig. 3). The experiment was designed in a way that simulates process such as torrential floods or runoff. Ten grams of crushed sample was immersed in 100 ml of distilled water and shaken for 24 h. Without drying periods, this time can correspond to 96 15-min-long events. Since the material used in the experiment was finely crushed the surface area was much larger and consequently the reaction was greatly enhanced. After shaking, material was filtered and contents of major and trace elements were determined in the leachate by Inductively coupled plasma – optical emission spectroscopy. The procedure used in this study is based on the procedure done by Faulkner et al. (2004).

Sodium adsorption ratio was calculated using the following equation (USDA, 1954):

$$\text{SAR} = \text{Exchangeable } \{(\text{Na})/(\text{Ca} + \text{Mg})^{0.5}\}$$

Where sodium, calcium and magnesium are in milliequivalents per liter (meq/l).

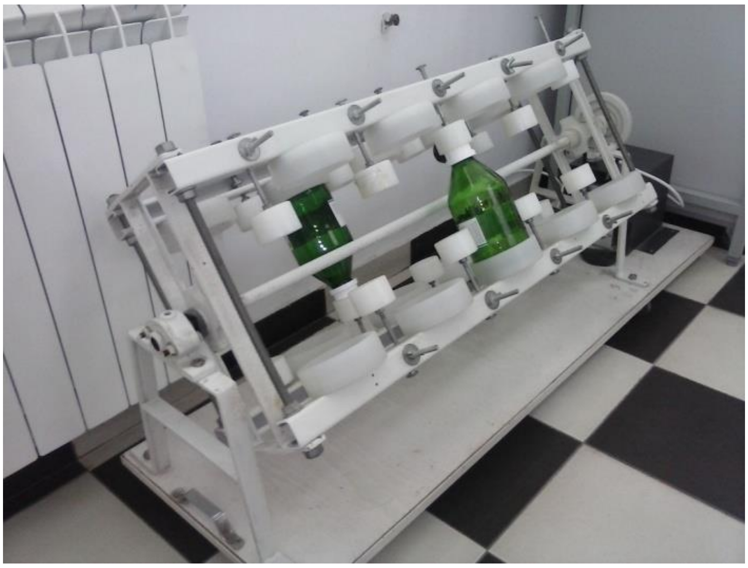

Fig. 3. Apparatus for leaching experiment

Results were statistically analyzed using the IBM SPSS Statistics 20 software. Data were analyzed using non-parametric Kolmogorov Smirnov test, correlation, and box plots.

RESULTS AND DISCUSSION

The lacustrine sediments in the Kremna Basin are represented by carbonates, marls, tuffs and tuffaceous sediments, and one characteristic layer of oil shale (Perunović et al., 2014). Outcrops are rare and can only be found at the basin edges. Surface of these outcrops are covered in irregular shards of various size indicating the dominance of physical weathering (Fig. 4). However, to precisely assess potential behavior of these sediments once they are exposed to surface conditions, their detail mineralogical and geochemical characteristics were determined. This is important since it has been proven that site physico-chemical properties and sediment and soil geochemical characteristics have a central role in geomorphological processes (Bryan, Yair, 1982; Imeson, Verstaten, 1988) and potential stabilization of dispersive materials (Faulkner et al., 2000; Faulkner, 2013).

Fig. 4. Physical deterioration of Kremna basin sediments

The Kremna basin carbonates are built of calcite and dolomite, while marly carbonates have varying amounts of clay (Fig. 5). Tuffs are built of searlesite, feldspar, dolomite, mica and chlorite, but some layers also contain monazite (Figs. 5 and 6). Presence of monazite indicates acid and alkaline volcanism (Dill, 2010). Oil shale is present in one characteristic layer and it is composed of smectite, dolomite and kaolinite as dominant minerals (Fig. 5).

Chemical composition of serpentinite and sediments (Table 1) is in accordance with their mineralogical composition (Fig. 5). Carbonates have high contents of CaO (30.04%), and MgO (10.52%) and lowest contents of K_2O (0.32%) and Na_2O (0.07%) of all tested sediments which is in accordance with the mineral composition since they are built mainly of dolomite. High content of MgO (27.55%) in marly carbonate is due to the presence of magnezite.

Concentrations of all tested trace elements are expectedly highest in serpentinite due to mineral composition of these rocks. Furthermore, the fact that mineral oxide surface sites of metal ions are largely bound to adsorbed carbonate minerals (Van Geen et al., 1984), or to natural organic matter (Redman et al., 2002), explains the concentrations of tested minor elements in carbonates, marls and oil shale.

Content of organic carbon (Corg) is higher in carbonates (5.33 %) and oil shales (4.34 %) than in marls (1.36 %) and tuffs (0.59 %). On the contrary, electrical conductivity (EC) is higher in marls (1128 µS cm^{-1}) and tuffs (846.50 µS cm^{-1}), than in carbonates (491.95 µS cm^{-1}), oil shales (552.60 µS cm^{-1}) and serpentinites (239.00 µS cm^{-1}). This is a clear indication of the presence of soluble minerals in marls and tuffs. Sodium adsorption ratio (SAR), as a dispersivity index, is high in marls (18.84) and tuffs (4.20). SAR for carbonates, oil shales and serpentinites is below 1 indicating low dispersivity.

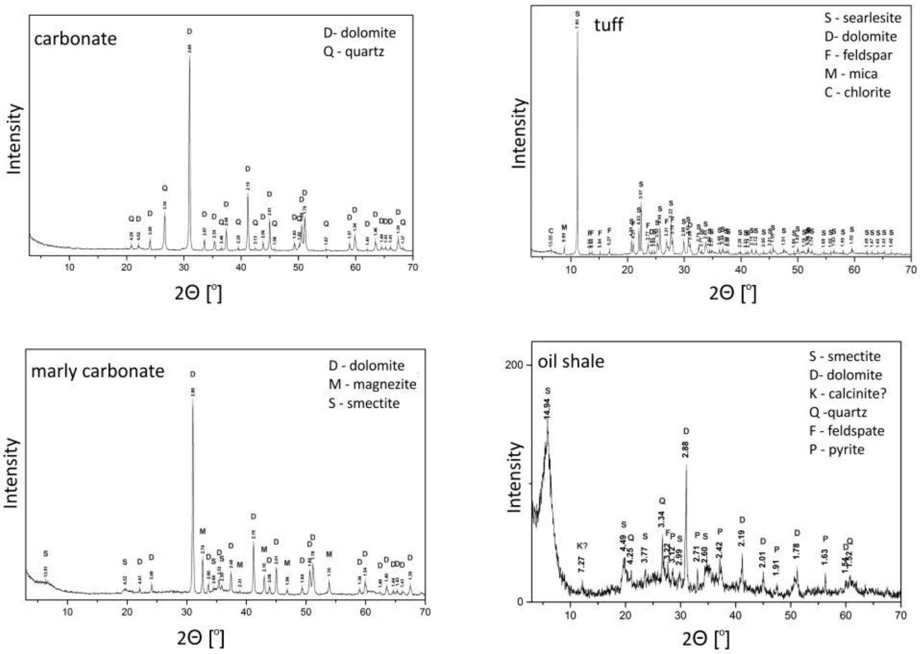

Fig. 5. X-ray diffractograms of tested sediments

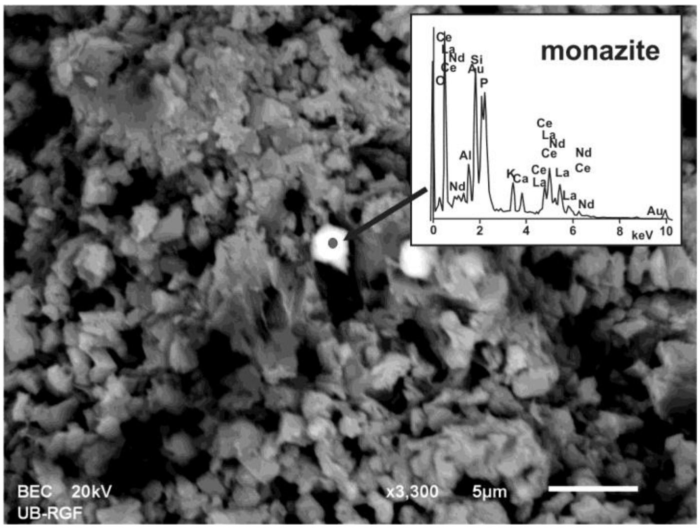

Fig. 6. SEM image of tuff

Table 1

Content of major and minor elements and physico-chemical properties of sediments from the Kremna basin

Sediment type		CaO (%)	K_2O (%)	Mgo (%)	Na_2O (%)	As (ppm)	Co (ppm)	Cr (ppm)	Cu (ppm)	Ni (ppm)	Pb (ppm)	SAR[a]	pH	EC[b] (µS cm^{-1})	Corg[c] (%)
carbonate	Av.	30.04	0.32	10.52	0.07	31.38	3.88	16.71	5.28	58.84	1.77	0.09	7.75	491.95	5.33
	St. dev.	7.36	0.22	5.53	0.03	13.62	2.08	9.97	2.29	29.55	0.71	0.04	0.74	275.80	2.66
tuff	Av.	7.02	0.70	17.67	1.09	5.10	19.64	131.10	29.09	362.42	3.38	4.20	9.21	846.50	0.59
	St. dev.	7.78	0.91	11.57	0.80	4.33	7.47	29.16	17.76	62.72	0.97	1.90	0.74	386.24	0.74
marly carbonate	Av.	7.41	0.68	27.55	1.06	4.22	23.06	158.63	5.96	454.79	2.85	18.84	9.39	1128.60	1.36
	St. dev.	3.48	0.82	8.25	0.60	2.64	11.22	80.29	2.92	249.54	2.94	13.32	0.21	639.83	0.89
oil shale	Av.	2.20	0.87	2.30	0.85	8.96	49.56	287.49	27.46	680.23	39.45	0.40	8.04	552.50	4.34
	St. dev.	0.17	0.17	0.45	0.04	0.59	3.18	15.51	1.64	33.38	4.20	0.02	0.01	17.68	0.15
serpentinite	Av.	0.24	0.02	24.93	0.96	7.87	86.53	297.88	98.17	1297.99	41.94	0.26	7.82	239.00	0.00
	St. dev.	0.06	0.01	0.93	0.11	0.80	8.10	11.23	2.06	107.40	2.65	0.11	0.06	12.73	0.00

[a] Sodium adsorption ratio’ [b] electrical conductivity, [c] organic carbon

During the leaching experiment highest amounts of sodium and potassium were leached from marls, sodium and magnesium from tuffs, while calcium was leached least of all tested elements (Fig. 7a). Losses of trace elements were proportionally greatest from tuffs suggesting that weathering of these sediments release highest concentrations of these elements into the environment (Fig. 7b).

The losses of K, Na and Mg do not show correlation with the total amounts of elements. Correlation coefficient between total and leached K is 0.195, total and leached Na 0.455, and total and leached Mg -0.144. The absence of correlation indicates that none of tested elements are bound only to one easily soluble mineral. On the contrary, K, Mg and Na are besides clay minerals bound to other mineral: Na is constituent of analcime and searlesite, K of feldspars, Mg of magnesite and dolomite. Statistically significant correlation exists only between total and leached Ca (correlation coefficient 0.556) indicating that this element is mostly found in carbonate minerals (calcite and dolomite).

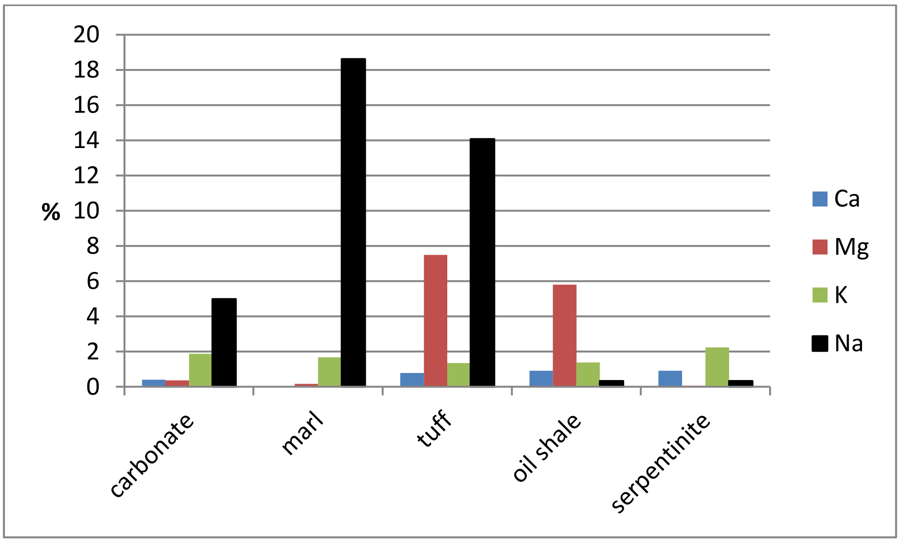

Fig. 7a. Leaching (%) of major elements

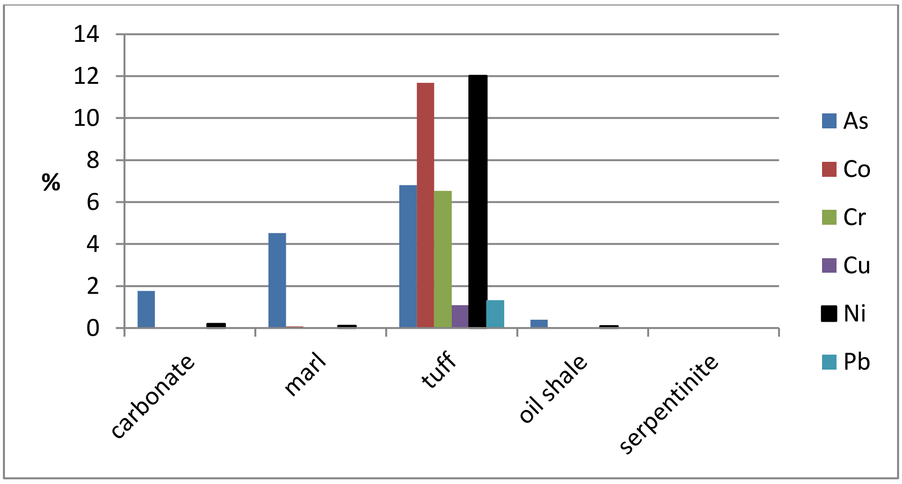

Fig. 7b. Leaching (%) of trace elements

Discussion

Dispersive properties of marls and mudstones were discussed by a number of authors (e.g. Lopez-Bermudez, Romero-Diaz, 1989; Faulkner et al., 2000; Kašanin-Grubin, Bryan, 2007 etc.). Very dispersive materials can enhance erosion processes (Bryan et al., 1978), and can slake and seal at very low rainfall intensities. Dispersion is usually measured by Sodium adsorption ratio (SAR).

In order to determine dispersive characteristics of tested sediments and serpentinite Sodium adsorption ratio (SAR), pH, electrical conductivity (EC), and content of organic carbon (Corg) were determined (Table 1). Rengasamy et al. (1984) used the SAR/EC ratio to determine the dispersive status of soils setting boundaries of the dispersivity domains. Based on this relation the authors differentiate dispersive, potentially dispersive and flocculated soils. The relationship between electrical conductivity (EC), and sodium adsorption ratio (SAR) is commonly used for description of dispersive materials (e.g. Naidu et al., 1995; Faulkner et al., 2000; Pulice et al., 2012).

Tested sediments for the Kremna basin generally fall into two large groups. Carbonates, oil shales and serpentinites are positioned in the field of potentially dispersive materials (Rengasamy class 2a), while marls and tuffs belong to the dispersive materials (Rengasamy class 1) (Fig. 8). The nonparametric Kolmogorov-Smirnov test confirmed that there is statistically significant difference between dispersive and potentially dispersive materials regarding the concentration of leached Na, SAR, EC, pH and Corg (Table 2).

Li et al. (2014) found that forest reclamation is a primary source of carbon emission, the secondary source is grassland cultivation and the tertiary source is shrubland and wetland reclamation. The concentration of organic matter increases rapidly with the vegetation coverage change in the surface soil layer (0-20 cm) when land coverage is above 60%, and decreases on a large scale when land coverage is below 30% (Wang et al., 2003).

The obtained difference occurs due to the presence of clay minerals in marly carbonates and tuff from which ions are easily released. Although the range of sodium concentrations is up to 2% in all tested samples (Table 1), leaching of Na in marly carbonates and tuffs on average is more than 25 time higher than in carbonates, oil shales and serpentinites (Fig. 7a). This means that erosion processes could be enhanced if marls and tuffs would get exposed to the surface conditions.

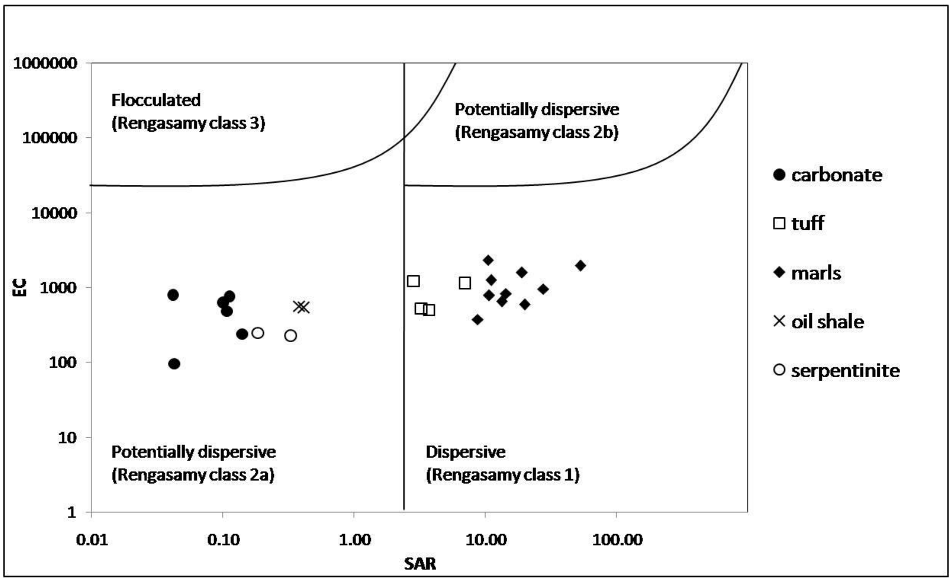

Fig. 8. Correlation between sodium adsorption ratio (SAR) and electrical conductivity (EC) for samples. (N.B. Data plotted on log transformed axes. Boundaries are given by Rengasamy et al., 1984, cited in Faulkner et al., 2000)

Table 2

Kolmogovor-Smirnov test for selected parameters between dispersive and potentially dispersive materials from the Kremna basin

Null hypothesis	Significance	Decision
The distribution of available Ca is the same across categories	0.175	Retain the null hypothesis
The distribution of available K is the same across categories	0.91	Retain the null hypothesis
The distribution of available Mg is the same across categories	0.91	Retain the null hypothesis
The distribution of available Na is the same across categories	0.00	*Reject the null hypothesis*
The distribution of SAR is the same across categories	0.00	*Reject the null hypothesis*
The distribution of EC is the same across categories	0.16	*Reject the null hypothesis*
The distribution of pH is the same across categories	0.00	*Reject the null hypothesis*
The distribution of Corg is the same across categories	0.00	*Reject the null hypothesis*

Generally, oil shale and marls from the Kremna basin have very similar mineralogical composition (Fig. 5). However, marls are very prone to dispersion, while oil shale did not show dispersive characteristics (Fig. 8). Reason for this might be the presence of organic matter, since content of organic carbon in oil shale is higher than in marly carbonates (Fig. 9, Table

1). In favor of this assumption is the fact that carbonates, as less dispersive materials also have high higher content of organic carbon than marls and tuffs. The box plot analysis was used to compare distributions of SAR and Corg in potentially dispersive materials and dispersive materials from the Kremna basin (Fig. 9).

Potentially dispersive materials, carbonates and oil shales, have higher contents of Corg, and dispersive materials (marls and tuffs) for SAR. Medians are also in different position indicating difference in distribution of parameters between the two groups. The observation is consistent with fact that immature organic matter, rich in functional groups has great affinity to mineral matter (particularly clays) of the sediments forming stable and complex aggregates. Moreover, numerous functional groups present in organic matter link metal cations forming the stable compounds (complexes and salts). However, small number of samples is not enough to draw firm conclusions, but this is certainly an indication for future research.

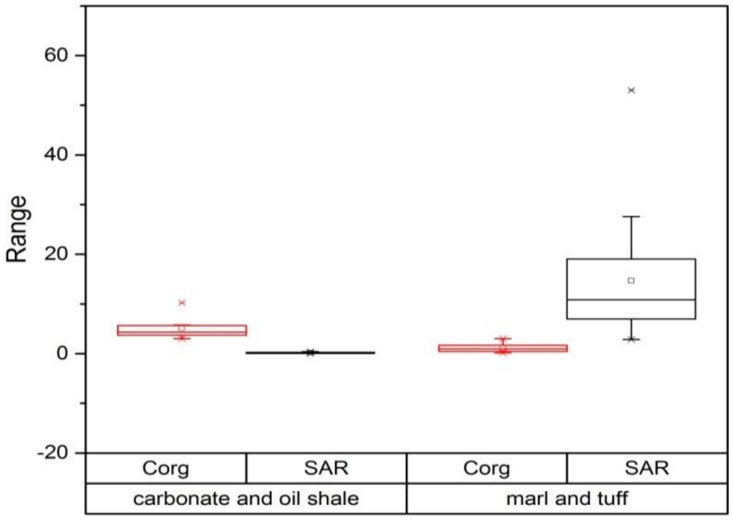

Fig. 9. Box plot of SAR and Corg for tested sediments

Weathering of ultramafic rocks in a range of climates, from arid to humid, can produce soils and clay minerals with a broad range of ion-exchange properties. Leaching of elements from serpentinites is strong, and elevated concentrations of Cu, Cr, Co and Ni in high to very high concentrations compared to average data for world soils, can be found soils overlying ultramafic rocks (Alexander, 2014). However, leaching experiment performed for the purpose of this study, which was designed to simulate at least 96 short precipitation events, showed that under those conditions leaching of heavy metals does not occur.

CONCLUSIONS

The Kremna basin is a typical lacustrine basin with abundant occurrences of carbonates and marly carbonates. Volcanic activity at the deposition time is evident through several layers of tuffs. Climate data indicate that both mean annual temperatures and precipitation have been showing more variability in recent decades, meaning that precipitation is usually more intense after dry periods. Such changes would enhance erosion process once they commence. Any change in land cover and land use would probably enhance erosion processes.

According to the results obtained in this study, tested sediments belong to two main dispersion categories: dispersive and potentially dispersive materials. Potentially dispersive are carbonates and oil shales, while marls and tuffs are dispersive materials, meaning they are prone to weathering, meaning that if these sediments were exposed to surface conditions they would undergo weathering and dispersion. Therefore, it can be concluded that the Kremna area is under slight to moderate hazard if a land use change would occur.

This study has proven that besides topographic characteristics, climatic data, and soil properties, detail mineralogical and geochemical composition should be determined prior to any land use change. Determination of sediment dispersion properties and content of organic matter is a good indication of material characteristics in a sense of predicting their behavior if they would get exposed to surface conditions.

Acknowledgments

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