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Mineralogical discrimination of the pleistocene loess/paleosol sections in Srijem and Baranja, Croatia

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ABSTRACT

Previous investigations of the mineralogical composition of loess sections (loess, loess-like sediments, paleosols, alluvial intercalations) in the Carpathian Basin have concluded that the Danube River is the dominant control on the loessitic parent material. These investigations also identify a significant role for the Danube's tributaries in creating local variations. The north–south alignment of these sections forms a transect from the central part of the Carpathian Basin to its southern edge. In this work, the mineral origin of loess sediments was identified by using the multivariate statistical method of discriminant function analysis. Two models were constructed based on the modal composition as the suite of predictor (independent) variables: one is using geographic location as the a priori grouping criterion (SECTION); another employing the difference between the sampling media (LITHOLOGY). Both of the examined discriminant models demonstrate the existence of the mixing zones. The Erdut section is a clear mixture of the mineralogies at the other studied locations, while loesses appear generally intermediate in mineralogy between alluvium and paleosol. The main rationale for the observed difference in modal composition between the Šarengrad and other analyzed sections is the proximity of the Šarengrad section to the Sava River floodplain and Dinaric Ophiolite Zone (DOZ), both important source areas for aeolian sediments in the southern edge of the Carpathian Basin that transport material from the Central Bosnian Mountains unit of DOZ. Chemically, the most resistant heavy minerals together with opaque minerals are exclusively associated with paleosols, being typical products of geochemical pedogenic processes.

1. Introduction

Since Gorjanović-Kramberger (1912, 1914, 1922) published his first results of loess investigations in Eastern Croatia, a number of recent studies – mostly paleontological (Banak et al., 2012; Hupuczi et al., 2010; Molnár et al., 2010) and mineralogical (Galović et al., 2009, 2011; Galović, 2014, 2016; Banak et al., 2012, 2013; Wacha et al., 2013), focused on loess sediments in this part of the Pannonian Basin. Simultaneously, in order to identify the provenance of material and local influences, a great number of modal analyses of loess in the Carpathian Basin was performed, initially in the pioneering work of Mutić (1975, 1989, 1990, 1993) and followed by a number of recent papers (Bronger, 2003; Marković et al., 2012, 2015; Thamó-Bozsó and Kovács, 2007; Thamó-Bozsó et al., 2014; Újvári et al., 2010, 2014). All these investigations confirmed the Alpine origin of Quaternary sediments, alongside of local influences. However, they also determined that the loess's generally homogeneous mineral content and uniform appearance successfully mutes any slight differences in modal composition.

The scope of this research includes the application of a multivariate statistical method with the purpose of recognizing the potential of differentiation among loess, paleosol and alluvial sediments from four loess sections of Eastern Croatia (based on the modal mineralogy dataset of previously collected 110 samples). Until now, these sections were studied in detail using geochronological, sedimentological, geochemical, mineralogical and paleontological methods (Galović et al., 2009, 2011; Galović, 2014, 2016). Using those methods, the evolutions of the Zmajevac I, Zmajevac, Erdut and Šarengrad sections were elucidated by defining the intensity and the chronological frames of climate changes. In the present case, multiple discriminant analysis (MDA) is used in order to explore the presence of possible patterns characterizing the modal composition of the analyzed sections. Application of MDA in the modal mineralogy is not a novelty (e.g., Eynatten et al., 2003; Heidke and Miksa, 2000), even in the neighboring areas and similar geological settings (Peh et al., 1998). However, lately it has only been used for the purpose of loess-like materials (Thamó-Bozsó et al., 2014). In the present work, a discriminant

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model is built as a tool for multiple group discrimination between loess sediments of the same origin (source area), regional correlation and characterization of mass movements. Also, the study is aimed at finding out whether a specific mineral composition is characteristic for certain sedimentary cycles during the Pleistocene and the degree of soil development in analyzed horizons, or if it is caused by the location of the analyzed sections with regard to the geological setting of the Carpathian Basin.

2. Location and geological setting of the sections

Four analyzed sections (Zmajevac, Zmajevac I, Erdut and Šarengrad) are located along the eastern border of Croatia along the Danube River (Fig. 1), in the area which is characterized by the temperate continental climate with dry summers (Peel et al., 2007). Their location, geological setting and stratigraphy are presented in detail in Galović et al. (2009, 2011) and Galović (2014, 2016) (Fig. 2). Generally, the Zmajevac I section consists of three paleosols, a laminated horizon and two loess layers; the Zmajevac section is built of four paleosols (one is a double paleosol), a laminated horizon and five loess layers; the Erdut section consists of four paleosols, a laminated horizon and five loess horizons, while the Šarengrad section consists of four paleosols, laminated horizon and three loess horizons (Galović et al., 2009, 2011; Galović, 2014, 2016) (Fig. 3).

2.1. Sampling and sample preparation

After removing a half meter of the outcrop in order to reduce the influence of weathering and vegetation, more than hundred horizons have been defined based on field observations (colour, grain size, structure, texture, bioturbations, presence and form of carbonates, etc.). In the final analysis, the total of 110 horizons were described (Galović et al., 2009; Galović, 2014) and samples were collected from the loess, paleosols and alluvial sediments at the four different sections to determine their mineralogical composition (Table 1).

Samples were air-dried for approximately one month. After drying, the samples were sieved to the <2 mm fraction to separate the sediment from larger carbonate concretions, while smaller, occasionally, remained in the samples (Galovic et al., 2011).

3. Modal composition of analyzed sections

To determine the qualitative and semi-quantitative mineral composition of heavy and light mineral associations, all samples were extracted after disaggregation in an ultrasonic bath and sieved to the 0.09–0.16 mm size fraction. It was then followed by dissolution of calcite. This fraction was selected for the analysis because it includes all virtual mineral species in proportions representative for the bulk sample. The heavy mineral fraction (HMF: opaque minerals (Op), chlorite (Chl), biotite (Bt), epidote–zoisite (Ep-Coe), amphibole (Am), pyroxene (Px), garnet (Grt), kyanite (Ky), staurolite (St), tourmaline (Tur), zircon (Zrn), rutile (Rt), titanite (Ttn), apatite (Ap), chromite (Chr)) was separated using bromoform ($CHBr_3$) at a density of 2.85–2.88 g cm^{-3}. Slides of the heavy and light mineral fraction (LMF: quartz (Qtz), feldspar (Fsp), muscovite (Ms), transparent lithic particles (LF)) were examined in polarized light. Qualitative and semi-quantitative composition of a sample was established after the determination of 300–400 grains and the percentage of each mineral was calculated. Canada balsam was used as the mounting medium.

Results of modal analysis are presented in Galović (2016).

4. Data processing

4.1. Compositional data and log-ratio analysis

A suite of 15 minerals, including the light and heavy mineral fraction, defined as an output of the modal analysis, was selected as predictor variables in building of the discriminant function model. The analyzed dataset consists of 31 loess, 11 alluvium and 68 paleosol samples collected from four loess/paleosol sections in Eastern Croatia, making 15-part mineral compositions of 110 samples altogether. Descriptive statistics (minimum, median and maximum) by a grouping variable (defined later in the text) for the entire dataset prior to data transformation is summarized in Table 2. This information is relevant if one is interested in relative values rather than absolute such as, for example, in the case of comparing similar investigations. However, the modal composition represents the classical example of compositional data (CoDa) in mineralogy, where correlations between relative abundances are problematic to interpret in absence of any other information or assumptions (Lovell et al., 2015). The nature of CoDa involves a mathematical property that all variables (compositions) in the analyzed sample sum to a unit value, usually expressed in percentages or mg/kg. As a result, all mineralogical, geochemical, and other datasets in geosciences are heavily plagued by the constant-sum constraint (CSC), creating a problem that interferes with procedures of conventional statistics. Original data represent parts of a whole, or fractions of a constant sum following geometry different from Euclidean (for details see, for example, Egozcue and Pawlowsky-Glahn, 2006), which is why they cannot fluctuate independently (closed data) and so produce the spurious correlations between compositions. Formally, CoDa cannot be represented in their raw form as points in the open, Euclidean space, where the scale is absolute, not relative. They refer to a restricted sample space known as simplex (simplicial complex) consisting of D parts or compositions (e.g. modal dataset). Thus a D-part composition (S^D) is really a subset of D-dimensional real space (R^D) (Pawlowsky-Glahn and Egozcue, 2006), which can assume the Euclidean vector space structure only after the appropriate transformation of its components. From several transformations presented in literature the centered log-ratio (clr) of raw (compositional) data, originally proposed by Aitchison (1986), is used in this work. The application of clr coefficients is considered essential in multivariate statistical analysis such as MDA as it preserves original distances between corresponding compositions (Egozcue and Pawlowsky-Glahn, 2006; Tolosana-Delgado, 2012). The problem of singularity innate to clr-transformed covariance matrix can be easily evaded by MDA working on the reduced data matrix, i.e. not resting on a full rank covariance matrix (Daunis-i-Estadella et al., 2011). This means removal of at least one composition (variable) after transformation. Since clr-transformed data represents unbounded real vectors in a real space, Mahalanobis distances (MD) stay invariant regardless of which component may be removed from the analysis (Barceló-Vidal and Pawlowsky-Glahn, 1999). Nonessential clr-transformed variables may be amalgamated and removed from further analysis.

Clr-coefficients can be computed from the following expression:

$$\mathrm{clr}\ (x) = \left(\log\frac{x_1}{g(x)}, \log\frac{x_2}{g(x)} \cdot \log\frac{x_3}{g(x)}, \ldots, \log\frac{x_D}{g(x)} \right) \quad (1)$$

where $x_1, x_2, x_3, \ldots x_D$ represent parts (compositions), and $g(x)$ represents the geometric mean of the parts. Calculated clr-variables are dimensionless numbers (ratios) that cannot be cross-compared directly and serve only as input data for MDA.

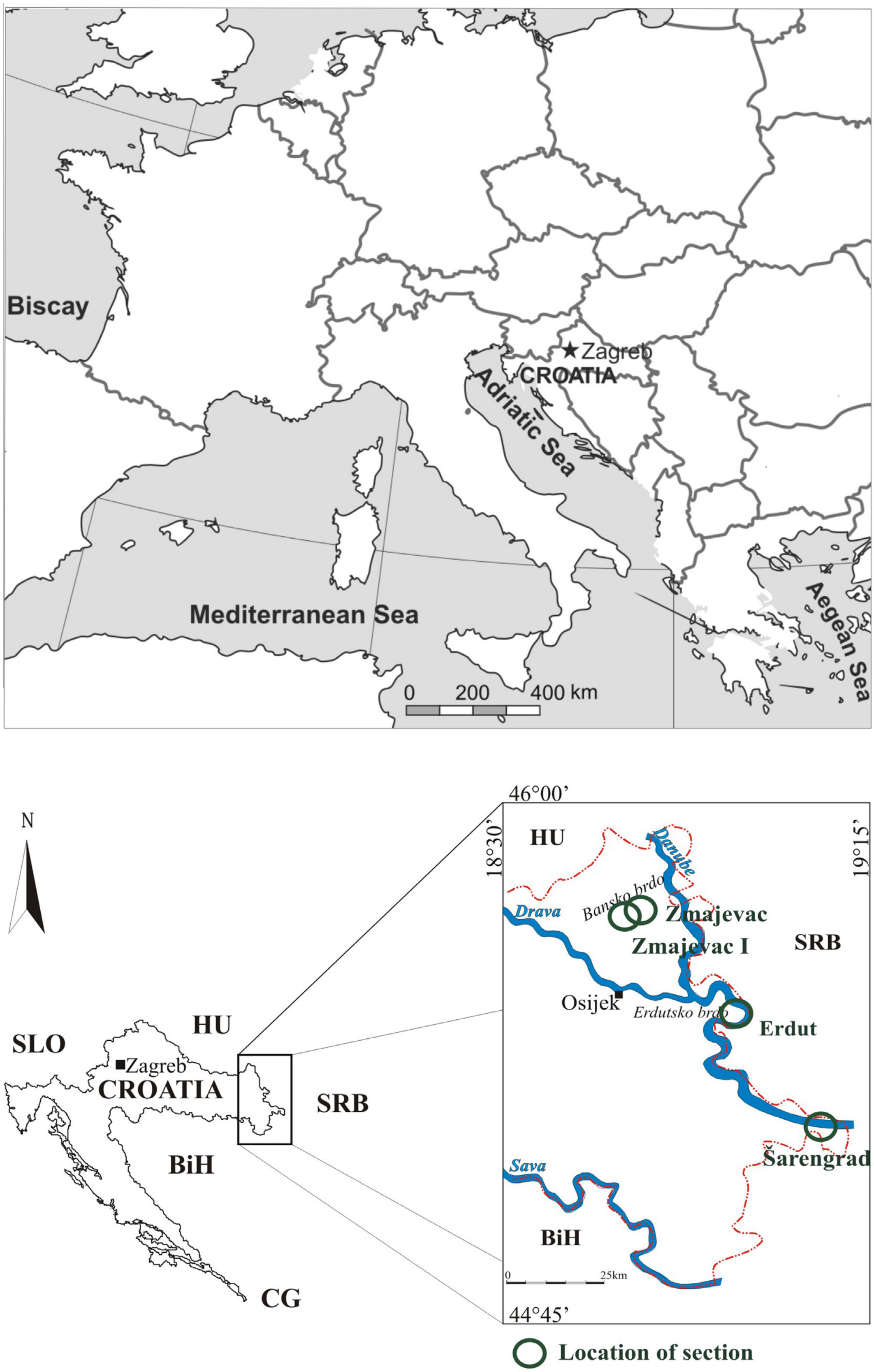


Fig. 1. Map showing the position of the sections under the investigation in the Eastern Croatia.

The feature stated above emerges from the very logic of the logratio analysis approach and deserves an additional piece of information concerning the algebraic-geometric structure of the sample space in order to follow results of the analysis in this work more easily. As explained by Aitchison and Egozcue (2005), compositional parts exhibit a twofold character: they may be displayed

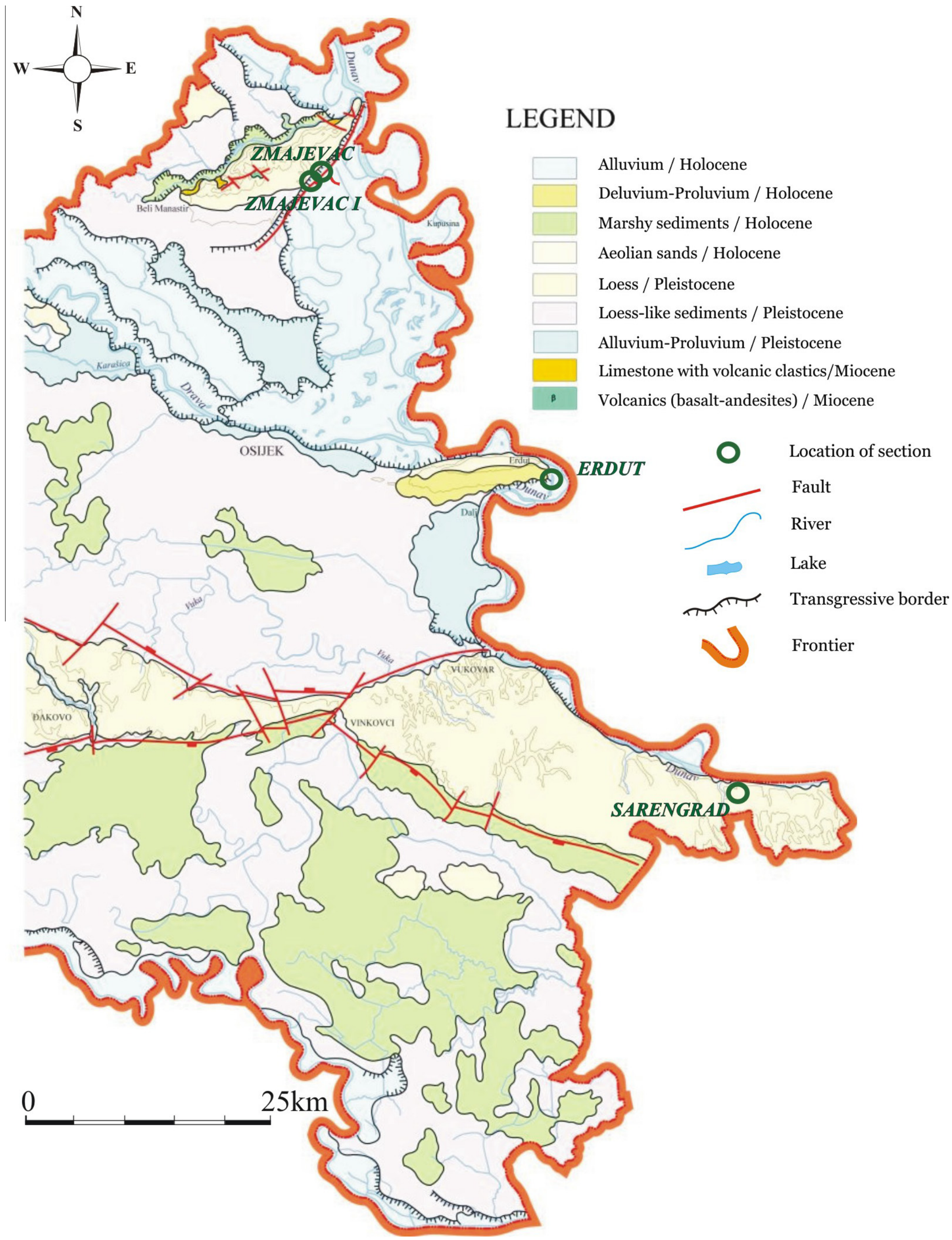


Fig. 2. Geological map of the Eastern Croatia (CGS – Department for Geology, 2009) showing the position of the sections.

either as: (a) raw compositional data such as percent values of the mineral (modal) compositions, using vectors of parts; or as (b) coordinates (scalars) in the Cartesian (orthonormal) coordinate system with Euclidean metric. However, in the latter case (the logratio analysis approach) they are not observed, de facto, as transformations of original data (compositional parts) such as, for example, simple log-transformations used in classical statistical techniques with purpose of data normalizing, but as coordinates. It is essential that these coordinates (clr coefficients) can be mapped onto orthogonal axes such as discriminant functions which are at 90°, thus forming a plane (two DFs) or three dimensional Euclidean space with three DFs. Mapping of original (raw) compositions directly on either coordinate scatterplots or biplots can easily lead to spurious results. One of the most obstinate fallacies associated with the classical (non-compositional) statistical methods arise from the situation that CoDa may sometimes simply confirm what is already well established through the application of traditional methods. However, the great caution must be exercised in this case since "...either we were lucky with our traditional methods, or at least the new methodology must be correct in this case" (Aitchison, 2008). Mark that clr-coefficients are computed by dividing the component parts (e.g. mineral percentage) by the geometric mean of all parts included in the analysis and finding their logarithm in the last analysis (Aitchison, 1986), as shown in Eq. (1).

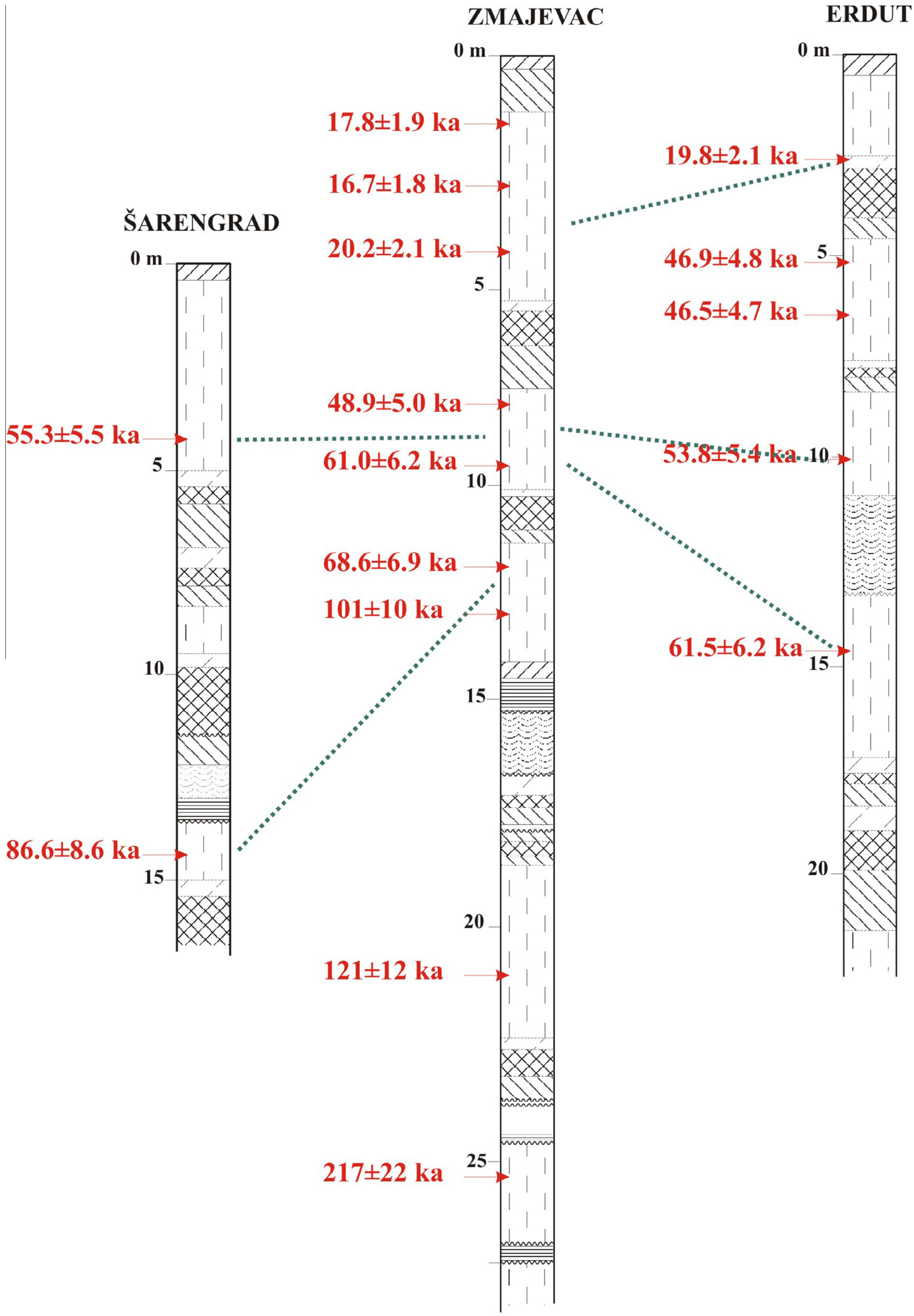


Fig. 3. Luminescence dating results of the Šarengrad, Zmajevac and Erdut sections (retrieved from Galović et al. (2009)).

4.2. Building the models

MDA is a conventional multivariate statistical technique, which is particularly useful in creating predictive models of multiple-group discrimination, based on the set of independent (predictor) variables. This method is frequently exploited in geosciences in cases where geological reasoning calls for some separation criterion independent of the variables from the analyzed dataset. In this

Table 1
List of samples, sampling depths and their lithology.

Zmajevac			Erdut			Zmajevac I			Šarengrad		
Sample	Depth (cm)	Hor	Sample	Depth (cm)	Hor	Sample	Depth (cm)	Hor	Sample	Depth (cm)	Hor
Z-1	130–330	Loess	E-1	200–247	Paleosol	ZI-1	30–125	Paleosol	Š-1	400–500	Loess
Z-2	400–520	Loess	E-2	247–278	Paleosol	ZI-2	125–165	Paleosol	Š-2	500–526	Paleosol
Z-3	520–570	Paleosol	E-3	278–307	Paleosol	ZI-3	165–185	Paleosol	Š-3	526–543	Paleosol
Z-4	570–595	Paleosol	E-4A	307–350	Paleosol	ZI-4	185–245	Paleosol	Š-4	543–581	Paleosol
Z-5	595–655	Paleosol	E-4B	350–400	Paleosol	ZI-5	245–281	Paleosol	Š-5	581–607	Paleosol
Z-6	655–675	Paleosol	E-5	400–450	Paleosol	ZI-6	281–305	Paleosol	Š-6	607–687	Paleosol
Z-7	675–775	Paleosol	E-6	450–530	Paleosol	ZI-7	305–315	Paleosol	Š-7	687–702	Paleosol
Z-8	775–990	Loess	E-7	700–750	Paleosol	ZI-9	330–370	Paleosol	Š-8	702–737	Paleosol
Z-9	990–1010	Loess	E-8	750–767	Paleosol	ZI-10	370–435	Loess	Š-9	737–780	Paleosol
Z-10	1010–1027	Loess	E-9	767–790	Paleosol	ZI-11	435–555	Loess	Š-10	780–830	Paleosol
Z-11	1027–1035	Paleosol	E-10	790–826	Paleosol				Š-11	830–944	Loess
Z-12	1035–1055	Paleosol	E-11	826–856	Paleosol				Š-12	944–959	Loess
Z-13	1055–1067	Paleosol	E-12A	856–906	Loess				Š-13	959–977	Paleosol
Z-14	1067–1090	Paleosol	E-12B	986–1076	Loess				Š-14	977–995	Paleosol
Z-15	1090–1105	Paleosol	E-13A	1080–1120	Alluvium				Š-15	995–1018	Paleosol
Z-16	1105–1135	Paleosol	E-13B	1120–1320	Alluvium				Š-16	1018–1057	Paleosol
Z-17	1135–1200	Loess	E-14A	1320–1370	Loess				Š-17A	1057–1082	Paleosol
Z-18	1200–1225	Loess	E-14B	1690–1720	Paleosol				Š-17B	1082–1142	Paleosol
Z-19	1225–1300	Loess	E-15	1720–1760	Paleosol				Š-18	1142–1214	Loess
Z-20	1450–1500	Alluvium	E-16	1760–1785	Paleosol				Š-19	1214–1294	Alluvium
Z-21	1500–1680	Alluvium	E-17	1785–1840	Paleosol				Š-20	1294–1352	Alluvium
Z-22	1680–1730	Alluvium	E-18	1840–1900	Paleosol				Š-21	1352–1361	Alluvium
Z-23	1730–1760	Paleosol	E-19	1900–1945	Paleosol				Š-22	1361–1492	Loess
Z-24	1760–1800	Paleosol	E-20	1945–1997	Paleosol				Š-23	1492–1512	Loess
Z-25	1800–1815	Paleosol	E-21A	1997–2009	Paleosol				Š-24	1512–1532	Paleosol
Z-26	1815–1840	Paleosol	E-21B	2009–2057	Paleosol				Š-25	1532–1550	Paleosol
Z-27	1840–1875	Paleosol	E-22	2057–2085	Paleosol				Š-26	1550–1620	Paleosol
Z-28	1875–1897	Loess	E-23	2085–2145	Loess						
Z-29A	1897–1957	Loess	E-24	2145–2200	Loess						
Z-29B	2047–2262	Loess									
Z-30	2262–2287	Loess									
Z-31	2287–2311	Loess									
Z-32	2311–2326	Paleosol									
Z-33	2326–2373	Paleosol									
Z-34	2373–2403	Paleosol									
Z-35	2403–2461	Paleosol									
Z-36	2461–2472	Loess									
Z-37	2472–2503	Loess									
Z-38	2503–2543	Loess									
Z-39	2543–2556	Alluvium									
Z-40	2556–2564	Alluvium									
Z-41A	2564–2664	Loess									
Z-41B	2664–2804	Loess									
Z-42	2804–2850	Alluvium									

work it is used in order to explore the differences in mineralogical (modal) datasets representing several loess/paleosol sections located in similar paleogeographical/neotectonic settings, typically prevailing in Eastern Croatia during the Middle and Late Pleistocene. The sections themselves, as well as the types of investigated media, or ‘‘lithology” (loess-paleosol-alluvium), represent the grouping criteria autonomous with regards to the observed modal compositions. These principles determine the strategy of analysis, which is conceptually based on building two different discriminant function models (DFM) (Rock, 1988).

In a statistical sense MDA is designed to maximize the between-group variance in comparison to the variance within each group, classifying each individual sample into one of the pre-defined groups with minimum error rate (proportion of misclassified objects, e.g. Davis, 1986; Dillon and Goldstein, 1984). A hypothesis is tested that all examined groups have the same multivariate mean against the alternative that at least one multivariate mean is different (Rock, 1988). Provided that the alternative hypothesis is not rejected, the original dataset is recalculated into discriminant scores assigning each object or group along one or more lines – linear discriminant functions. The multivariate problem is reduced thereby into a fewer-dimension solution, one less than the number of groups (K-1).

The scope of this study is focused on building predictive discriminant models with maximum classification efficiency and established on: (a) four a priori defined groups representing investigated sections from different geographic locations in Eastern Croatia – Zmajevac, Zmajevac I, Erdut and Šarengrad (irrespective of sample media) – SECTION model, and; (b) three a priori defined groups representing the sample media – loess, paleosol and alluvium (irrespective of sampling locations) – LITHOLOGY model. In both cases the same set of 15 clr-transformed compositions representing the results of modal analysis are included in the model – light mineral fraction (Qtz, Fsp, Ms, LF) and heavy mineral fraction (Op, Chl, Bt, EpCoe, Amp, Px, Grt, Ky, St, Tur, Zrn). Due to their insignificant presence (<0.5%) in the mineral composition, four minerals (Rt, Ttn, Ap, Chr) are amalgamated and discarded. To this end was the discriminant function analysis from the statistical software package of STATISTICA, Release 7.1 (StatSoft, Inc, 2006) used in order to achieve the best separation between the groups and explain the possible geological causes for the structure of input data by discriminant model.

Table 2
Descriptive statistics of raw (compositional) mineralogical data for SECTION and LITHOLOGY criteria.

Group	Zmajevac (44)			Zmajevac I (10)			Erdut (29)			Šarengrad (27)		
Mineral	Min	Med	Max	Min	Med	Max	Min	Med	Max	Min	Med	Max
(a) SECTION												
Qtz	6.79	56.68	80.90	62.10	72.99	76.96	26.06	56.24	72.28	19.08	53.31	77.96
Fsp	0.97	9.43	18.72	7.68	10.09	13.36	2.90	9.08	19.35	1.92	10.33	30.46
Ms	2.87	16.32	84.39	1.84	6.35	20.99	0.95	19.17	64.65	3.00	24.50	69.24
LF	0.97	6.16	14.59	3.84	5.73	11.10	1.88	5.82	19.63	0.98	3.85	13.99
Op	0.00	0.26	0.96	0.00	0.60	1.40	0.10	0.50	3.01	0.02	0.66	3.05
Chl	0.02	0.86	3.47	0.00	0.22	0.70	0.14	0.72	5.49	0.00	0.89	4.41
Bt	0.00	0.19	1.96	0.00	0.10	0.30	0.00	0.17	1.46	0.00	0.18	0.81
EpCoe	0.00	0.40	1.65	0.0	0.49	1.12	0.26	0.61	4.73	0.01	0.58	2.77
Am	0.00	0.48	2.05	0.00	0.91	2.27	0.26	0.85	3.74	0.00	0.33	2.50
Px	0.00	0.06	0.32	0.00	0.07	0.21	0.00	0.07	1.39	0.00	0.10	0.95
Grt	0.00	0.72	3.85	0.00	1.45	4.47	0.38	0.93	5.99	0.00	0.62	2.06
Ky	0.00	0.07	0.31	0.00	0.21	0.45	0.00	0.07	0.18	0.00	0.04	0.20
St	0.00	0.10	0.31	0.00	0.26	0.37	0.03	0.14	0.86	0.00	0.08	0.45
Tur	0.00	0.07	0.31	0.00	0.09	0.41	0.00	0.10	0.28	0.00	0.04	0.25
Zrn	0.00	0.03	0.12	0.00	0.11	0.30	0.00	0.05	0.58	0.00	0.03	0.24

	Loess (31)			Alluvium (11)			Paleosol (68)		
	Min	Med	Max	Min	Med	Max	Min	Med	Max
(b) LITHOLOGY									
Qtz	6.79	52.84	76.96	15.20	29.64	47.03	26.06	61.74	80.90
Fsp	1.92	9.07	16.19	0.97	6.86	19.35	2.90	10.33	30.46
Ms	1.88	20.99	84.39	7.57	52.92	79.38	0.95	11.64	64.65
LF	0.98	6.54	19.63	0.97	4.90	18.15	1.88	5.77	14.28
Op	0.00	0.37	3.01	0.00	0.25	2.01	0.02	0.49	3.05
Chl	0.00	1.05	5.49	0.03	2.10	4.34	0.00	0.57	2.11
Bt	0.00	0.26	1.96	0.00	0.42	1.65	0.00	0.13	1.13
EpCoe	0.00	0.51	2.25	0.01	0.35	3.36	0.01	0.55	4.73
Am	0.00	0.43	3.49	0.00	0.51	3.74	0.00	0.64	3.14
Px	0.00	0.07	0.56	0.00	0.11	1.39	0.00	0.07	0.95
Grt	0.00	0.71	3.85	0.00	0.43	5.99	0.00	0.76	4.47
Ky	0.00	0.04	0.31	0.00	0.01	0.11	0.00	0.07	0.45
St	0.00	0.08	0.57	0.00	0.05	0.42	0.00	0.11	0.86
Tur	0.00	0.05	0.31	0.00	0.06	0.28	0.00	0.08	0.41
Zrn	0.00	0.03	0.41	0.00	0.04	0.56	0.00	0.05	0.37

Note: minimum values less than 0.005% (Min < 0.005%) trimmed to 0.00%

Table 3
Multivariate test for overall significance of discrimination and tests of residual roots.

(a) SECTION model							
No. of variables				15			
Wilks' lambda				0.2267			
Approximate *F* ratio				3.947			
Degrees of freedom				[45; 274]			
p-level				<0.000			
DF	Eigen value	Eigen (%)	Canon. *R*	Wilks' λ	chi^2	df	*p*-level
1	1.369	64.39	0.760	0.227	147.7	45	0.000
2	0.574	27.00	0.604	0.537	61.8	28	0.000
3	0.183	8.61	0.393	0.845	16.7	13	0.212
(b) LITHOLOGY model							
No. of variables				15			
Wilks' lambda				0.4858			
Approximate *F* ratio				2.695			
Degrees of freedom				[30; 186]			
p-level				<0.000			
DF	Eigen value	Eigen (%)	Canon. *R*	Wilks' λ	chi^2	df	*p*-level
1	0.786	83.71	0.663	0.486	72.2	30	0.000
2	0.153	16.29	0.364	0.867	14.2	14	0.434

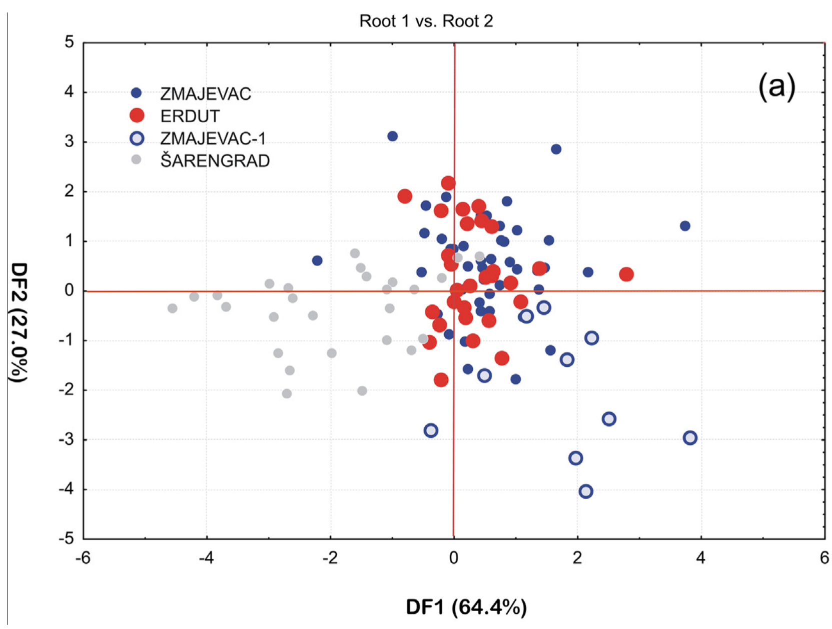
5. Results and discussion

5.1. Labeling discriminant space

Results of the MDA are summarized in the common table (Table 3) describing the two exploratory models. The table comprises the multivariate test for the overall significance of discrimination and the test of residual roots (discriminant functions) for both cases. The Wilks' λ statistical test is employed regularly in order to confirm the probability level ($p < 0.05$) safe to proceed with computing discriminant functions (DFs). It is also used to select the statistically significant functions explaining the maximum of the within-group variation. In all practical applications, after computing DFs the relationship between the variables and groups is displayed in separate scatterplots instead of biplots as their multivariate correlates. Application of scatterplots is compulsory, because the origin of the biplot no longer corresponds to the geometric mean of the dataset after eliminating the amalgamated variable (Rt + Ttn + Ap + Chr) in order to compensate for matrix ill-conditioning. Scatterplots are the straightforward tool essential in the procedure of labeling discriminant functions (affiliation between groups and variables) or transfiguration of functional (structural) model into a process (mineralogical) one. Based on discriminant loadings, the variable scatterplot explains the contribution of each DF to a single geologic process responsible for the separation. While the variables participate in this structure-process transformation as the building blocks assuming the role of process descriptors. Simultaneously, the individual objects (samples) and groups are represented on the scatterplot of discriminant scores. Interdependence between variables and samples in the reduced discriminant space is always interpreted using their corresponding position along the respective axis in both models.

5.1.1. The SECTION model

In the computed SECTION model, the first discriminant function DF1 makes by far the greatest contribution to discrimination between the groups, accounting for more than 64% of the total variability (Table 3). Together with DF2, it amounts to over 91%. On the other side of the scale is the third discriminant function DF3, which can be excluded from further consideration in virtue of the high statistical significance ($p = 0.21$). The latter considerably exceeds the limit of the acceptable error level (routinely $p < 0.05$), curtailing the reliability of discrimination beyond practical purpose. DF1 is bipolar and can be interpreted as reflecting inverse relationship between the mineral suite including Op, Px and Chl against another suite of minerals headed by kyanite (Ky). In this model, a close inspection into the related group and variable scatterplots (Fig. 4a against b) reveals that this pattern discriminates between the groups rather loosely, separating only the Šarengrad group from the rest, where Zmajevac I tends to branch out from Zmajevac. Essentially, the Zmajevac and Erdut groups occupy the central diagram area, conveying information of the average mineralogical composition with regard to the former two groups. The scatterplots reveal the tendency of enrichment of the Šarengrad group in Op-Px-Chl-(EpCoe) suite at the expanse of Ky-(Am) set characterizing the Zmajevac I and (partly) Zmajevac groups, and vice versa. Although statistically significant, discrimination between the groups is less than 73%, meaning that 80 out of 110 samples are classified correctly. It applies mostly to the Zmajevac I and Zmajevac groups, where only 20% is lost to the other groups (Table 4). On the other side, the Erdut group is immersed in the central cloud of objects losing almost half of its samples to the Zmajevac group (only 55% correctly classified). Correct assignment of Zmajevac I cases is determined largely by the second discriminant function DF2, precisely in the part that relates to samples enriched in minerals such as Zrn, Qtz and Fsp, while at the same time, minerals such as Chl and EpCoe are clearly deficient in this group. This arrangement is a potentially significant provenance indicator leading to felsic (granitic) igneous rocks as a possible parent material for this location, although some samples (including those from the Zmajevac section) may be enriched in kyanite. Going back to DF1, the polarity between the Op-Px-Chl-



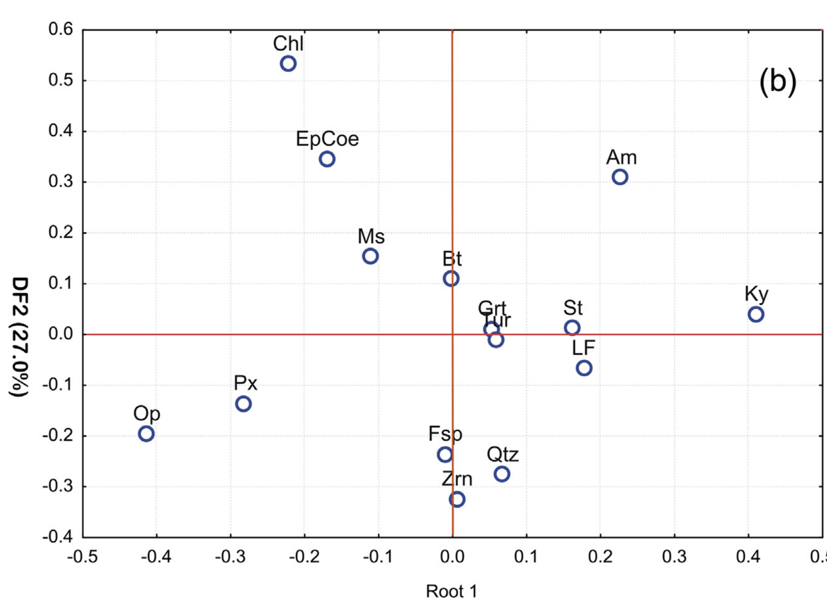


Fig. 4. Comparison between variables and groups in the SECTION CoDa DFM (clr-transformed data): scatterplots of (a) individual objects (samples), and (b) variable loadings in reduced discriminant space of the first two discriminant functions (DF1–DF2).

Table 4
Classification matrix.

Observed Groups	Predicted groups					
	Zmajevac	Erdut	Zmajevac I	Šarengrad	Total obs.	% correct
SECTION model						
Zmajevac	**35**	7	1	1	44	79.55
Erdut	13	**16**	0	0	29	55.17
Zmajevac I	2	0	**8**	0	10	80.00
Šarengrad	4	2	0	**21**	27	77.78
Total pred.	54	25	9	22	110	72.73

Observed groups	Predicted groups				
	Loess	Alluvium	Paleosol	Total obs.	% correct
LITHOLOGY model					
Loess	**17**	3	11	31	54.84
Alluvium	6	**4**	1	11	36.36
Paleosol	8	2	**58**	68	85.29
Total pred.	31	9	70	110	71.82

Note: Rows are observed classifications; columns are predicted classifications.

(EpCoe) and Ky-(Am) mineral assemblages may be interpreted in the provenance terms, as a contrast between the ultra-maphic igneous and medium- to high-grade metamorphic provenance, characterizing the Šarengrad (Px) and the Zmajevac I sections.

5.1.2. The LITHOLOGY model

This model is characterized by (still inferior) classification efficiency with respect to the previous case (Table 4). Although similar to the SECTION model in general terms (72%), this relatively high overall classification rate is due to the paleosol samples, which are classified correctly with more than 85%. The other two groups – loess and, alluvium in particular – are poorly classified. This unfavorable fact results from the fact that the single discriminant function is statistically significant, explaining almost 84% of the total variability in the analyzed data (Table 3). DF1 separates most of the paleosol samples (of the rest the greatest part was lost to loess) from alluvium, based on the Ky/Ms-Chl variable polarity (Fig. 5a and b). However, alluvium itself is not quite distinguished from loess – of which some samples are even richer in Ms-Chl suite with regards to alluvium. As well as in the SECTION model, kyanite in paleosol may reflect the input of material derived from ultramaphic (ophiolite?) and medium/high grade metamorphic rocks during the period of its formation. Recycled chlorite and muscovite (Chl-Ms) often derive from metamorphic mountain belts undergoing moderate to strong weathering (Potter et al., 2005). The overall picture (Fig. 5) points at loess as the central hub from which two branches separate in opposite directions – to the process brought to an end at some point in the past (paleosol), and to active process operating in the more or less recent times (alluvium). These processes resulted in materials with widely different modal composition, as can be seen from the group samples interchange. The alluvium group seems particularly unstable as majority of its samples are lost to the (central) loess group. Indeed, the closeness between loess and paleosol demonstrates that sedimentary and environmental processes in the study area must have been rather different in the past, since only one alluvium sample is confused with paleosol (Table 4). However, the loess-alluvium interplay indicates that most of alluvium in the investigated area originates from the previous loess deposits that had been eroded and redeposited by fluvial processes in the recent times, irrespective of the studied sections.

5.2. Origin of aeolian sediments

As already presented in numerous publications, loess derived from the floodplains of the Danube River and its tributaries is mostly derived from the Alps. Based on slight differences in composition of the heavy mineral fraction, local alluvial influence is demonstrated by Banak et al. (2013), Buggle et al. (2008, 2013, 2014), Galović (2014), Rubinić et al. (2015), Thamó-Bozsó and Kovács (2007, 2014) and Újvári et al. (2010). Galović (2016) emphasizes two important local influences: muscovite bearing rocks (Balen et al., 2006) and chlorite bearing rocks (Pamić et al, 2002), that fed alluvial accumulations – additional secondary source material for aeolian sediments. Insight into computed models corroborates these findings, especially with regard to the LITHOLOGY model, which appropriately classifies alluvium and part of the loess samples into the Ms-Chl dominated zone on the pertinent diagrams (Fig. 5). A truly effective mixing of influences is still more evident in the SECTION model, where the Erdut section appears as a focal point situated geographically almost exactly between the Zmajevac and the Zmajevac I sections to the north and the Šarengrad section to the south (Figs. 2 and 4). In a statistical sense, the Erdut section is a perfect example of an average mineralogical composition distributed in space (horizontally). In contrast, the LITHOLOGY criterion is concerned essentially with vertical disposition. That is the arrangement in time where the contribution of alluvium to the formation of loess is quite obvious from diagrams (Fig. 5) and classification ("confusion") matrix (Table 4). They disclose a massive samples exchange between the two groups, indicated by very low classification efficiency for both groups, alluvium in particular (36%), meaning that a solid portion of its samples had already gone through the transformation process in the relatively recent times. On the contrary, the paleosol group is relatively homogeneous (with 85% classification rate), with almost no affiliation to alluvium, and thus referring to processes completed before deposition of alluvial sediments. In this respect, muscovite and chlorite can indubitably be seen as a relatively modern contribution to the mineralogy of the investigated area as opposed to minerals such as kyanite that most properly represent more "ancient" paleosols (Fig. 5). Muscovite, chlorite and marginally biotite are associated with alluvial sediments intercalated into the investigated sections, probably associated to accumulation by fluvial flow, as a consequence of its specific platy shape. Coevally, kyanite is associated to paleosols, as this mineral is prone to physical weathering, and resistant to chemical weathering. Chemically the most resistant heavy minerals (kyanite, tourmaline, garnet and staurolite) are exclusively associated with paleosols, along with opaque minerals, most common products of geochemical pedogenetic processes.

Galović (2016) discusses that the heavy mineral association is similar to the Danube flood plain sediments (Thamó-Bozsó and Kovács, 2007) and loess from Hungary (Thamó-Bozsó et al., 2014), but the southern edge of the Carpathian Basin was very likely influenced by the nearby Dinaride Ophiolite Zone (Rubinić et al., 2015). Garnet, amphibole and epidote-zoisite are dominant minerals within the transparent heavy mineral composition (THM) of all samples, although their discriminant potential may not be particularly high. This applies especially to garnet which, in both models, falls close to the DF1/DF2 axis intersection (Figs. 4b and 5b). On this account, garnet seems quite unhelpful in distinguishing either between sections or horizons. This indicates the Danube floodplain region (Thamó-Bozsó and Kovács, 2007) and redeposited loess from Hungary (Thamó-Bozsó et al., 2014) to be the main source of the analyzed material. Since the Danube River originates from the same region as the Sava and the Drava Rivers (the Alpine region), their mineral compositions are similar. Buggle et al. (2008) geochemically characterized the loess/paleosol sections of Batajnica/Stari Slankamen (Serbia), Mircea Voda (Romania) and Stary Kaydaky (Ukraine) in order to identify the origin of south-eastern and eastern European loess deposits. They concluded that the Danube catchment area is the

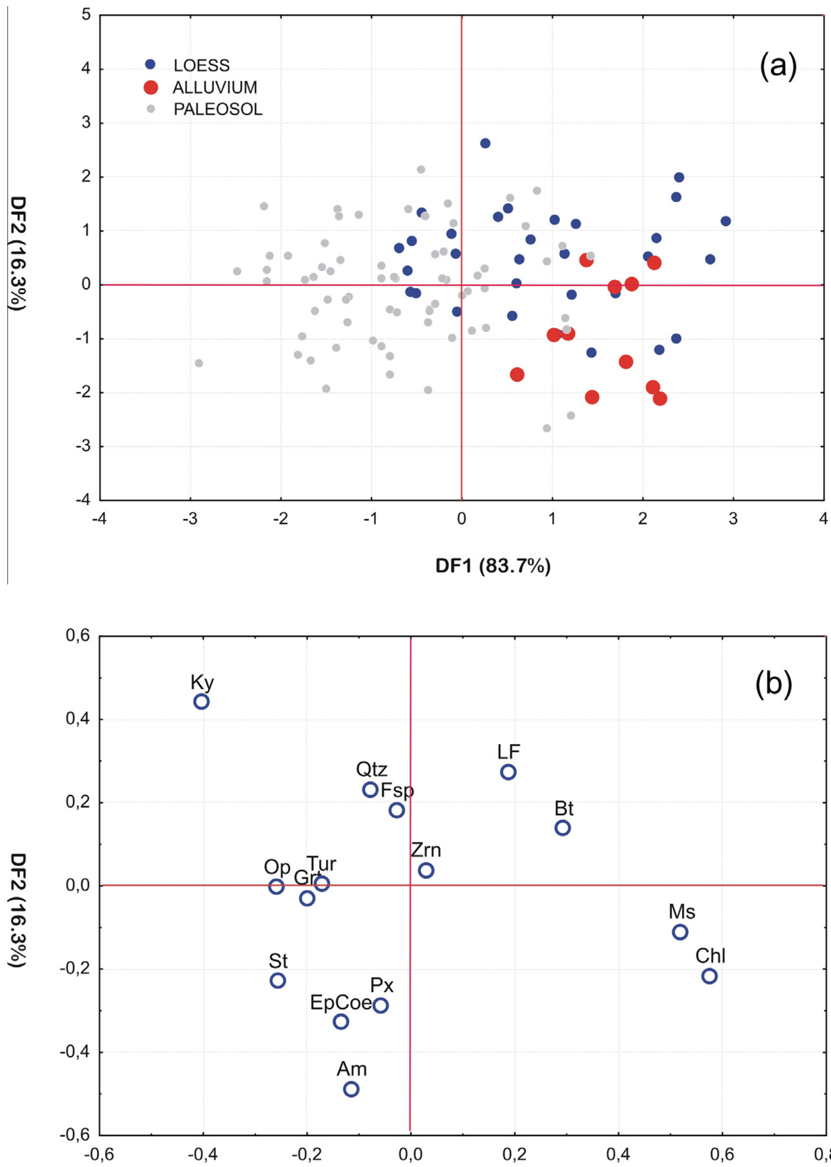


Fig. 5. Comparison between variables and groups in the LITHOLOGY CoDa DFM (clr-transformed data): scatterplots of (a) individual objects (samples), and (b) variable loadings in reduced discriminant space of the first two discriminant functions (DF1–DF2).

most important for the Pleistocene delivery of the silt-sized alluvial sediments in the area. They also considered the Drava and Sava rivers as additional important silt sources supplying glacio-fluvial sediments of the eastern Alps, with respect to the element composition and weathering products (Galović, 2014, 2016).

Analyzed sections are situated along the Danube River, forming a transect with a general course from the north to the south, and a length of approximately 30 km (Figs. 1 and 2). This transect has sub-meridian direction and it is perpendicular to the southern edge of the Carpathian Basin and to the contact of the Carpathian Basin and the Central Bosnian Mountains unit of the Dinaric Ophiolite Zone (DOZ) (Pamić et al., 2002; Robertson et al., 2009). The contact is marked by the Sava River waterway. Although the Sava River is the river of Alpine origin approximately only 50,000 years old, it collects in its lower part the material brought by Bosnian Rivers (the Una, the Bosna and the Vrbas Rivers representing the Sava River southern tributaries) eroding DOZ. Moreover, the Sava River also erodes Tertiary floodplain sediments formed by its Bosnian tributaries, because the erosion of Alpine ophiolites from the central and NW Dinarides (DOZ) started during the Alpine orogenesis following the uplift of the Dinarides (Pamić et al., 2002). DOZ is mostly composed of ore bearing mafic and ultramafic rocks, providing the alluvial material of the Sava River southern tributaries with pyroxenes, chlorite and opaque minerals (chemically unstable minerals). On this account, the Sava River floodplain, otherwise dominated by Alpine components, is enriched in the HM assemblage due to alluvial input from the Central Bosnian Mountains. Because of the short alluvial transport from the source area, these chemically unstable minerals are still preserved in the finally formed aeolian sediments of the southernmost Šarengrad section, before they are finally deposited as alluvial sediment. This is obvious from the diagrams of the SECTION model (Fig. 4), where the Op-Px-Chl mineral assembly coincides with the position of the Šarengrad section. Assessment of a locally-sourced loess system in Europe is presented on example of The Swiss Jura Mountains (Martignier et al., 2015). Furthermore, there are no significant differences in the content of pyroxene in loess, paleosol or alluvial sediments; neither portion of pyroxene is changing with depth (age) (Fig. 5). On the other hand, typical stable minerals, including tourmaline and metamorphic minerals such as staurolite, zircon, and especially kyanite (Schokker et al., 2005), are enriched in the Zmajevac I and the Zmajevac. These sections are located on the north of the transect and the farthest from the Sava River tributaries and their floodplains. Along with quartz, feldspars and zircon, they make the main characteristics of the Zmajevac I section (Fig. 4), situated farthest to the north of the investigated area (Fig. 2). The obvious discrimination between the Zmajevac and the Zmajevac I sections, which are close to each other (250 m), must be ascribed to predominance of paleosol horizons (profoundly distinguished by Ky, Fig. 5) over loess and alluvium on the spot. In general, significantly higher content of the light mineral fraction (LMF) in the Zmajevac I and the Zmajevac sections is in all probability the consequence of long distance fluvial transport (from the Alps to the south of the Carpathian basin), followed by intensive chemical weathering (water was the dominant transport media), since the LMF is composed of 50–75% of quartz grains (Galović, 2016; Újvári et al., 2010, 2014; Thamó-Bozsó and Kovács, 2007; Thamó-Bozsó et al., 2014). The main rationale for the observed difference in modal composition between the Šarengrad section and other analyzed sections can be found in the vicinity of the former to the Sava River floodplain and DOZ, figuring as important source areas for aeolian sediments in the southern edge of the Carpathian Basin.

Buggle et al. (2013) proposed that this trend is related to Quaternary surface uplift of European mountain ranges, specifically the Alps, Carpathians and Dinarides. Available paleoelevation studies for these mountain ranges as reviewed by Buggle et al. (2013) suggest several hundreds of meters of paleoelevation change during the Middle Pleistocene. Buggle et al. (2013) hypothesized that already such small scale changes in mountain topography might have a climatic impact (changes in atmospheric circulation, rain shadow effects) on leeward situated lowlands and basins as the middle and lower Danube Basin. This hypothesis is invoked as "mountain uplift hypothesis".

6. Conclusions

The comprehensive investigations of modal composition focused on loess sections (loess, loess-like sediments, paleosols, alluvial intercalations) in the Carpathian Basin have been performed with purpose of mineralogical fingerprinting of their parent material. In most cases, conclusions drawn from these investigations highlighted the dominating influence of the Danube River. Furthermore, they identified the local controls of its tributaries as well. The north–south alignment of sections forming a transect, from the central part of the Carpathian Basin to its southern edge, provided profound insights into the distribution of mineral composition. It showed the abating influence of the Danube River and an increased impacts of rivers draining the Central Bosnian Mountains of the Dinaric Ophiolite Zone (DOZ). In this work, mineral fingerprinting (based on modal composition) of loess sediment sources was carried out using the multivariate statistical method of discriminant function analysis as a highly specific tool, having the considerable differentiating potential. It identified the zones of mixing in both discriminant models: in the SECTION model it is reflected in the Erdut section; while in the LITHOLOGY model, the loess for its most part mediates between alluvium and paleosol.

A truly effective mixing of influences is most evident in the SECTION model where the Erdut section appears as a focal point, situated geographically almost exactly between the Zmajevac and the Zmajevac I sections (in the north) and the Šarengrad section (in the south). In a statistical sense, the Erdut section is a perfect example of an average mineralogical composition distributed in space. Significantly higher content of the LMF in the Zmajevac I and the Zmajevac sections is in all probability the consequence of a long distance fluvial transport (from the Alps to the south of the Carpathian basin). It was then followed by an intensive chemical weathering (water was the dominant transport media), whereas, in the Šarengrad section, higher content of the HMF indicates shorter fluvial transport. Thus, the main rationale for the observed difference in modal composition between the Šarengrad section and the other analyzed sections, can be found in the vicinity of the former to the Sava River floodplain and DOZ, figuring as important source areas for aeolian sediments in the southern edge of the Carpathian Basin and indicating the influence of the Central Bosnian Mountains unit of the Dinaric Ophiolite Zone (DOZ).

As for lithological discrimination between the sampled media (LITHOLOGY model), minerals such as muscovite, chlorite and, marginally, biotite are all associated with alluvial sediments intercalated into the investigated sections. It was most likely a result of accumulation by fluvial flow due to their specific platy shape. Coevally, kyanite is related to paleosols, as a mineral prone to physical, but resistant to chemical weathering. Chemically, the most resistant heavy minerals (kyanite, tourmaline, garnet and staurolite) are exclusively associated with paleosols, together with opaque minerals, the most often products of geochemical pedogenetic processes.

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