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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.

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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:

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

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.

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