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# Evidence and mineralogical and physico-chemical properties of chernozem and chernozem-like soils in Croatia

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## Abstract

The aim was to determine possible local differences between the parent materials of recent leoss-derived soils in eastern Croatia (Dalj, Zmajevac). Furthermore, it highlights the existence of chernozem and chernozem-like soils in Croatia and describes their basic physical, chemical and mineral properties. For this purpose, two soil profiles (P-3 and P-6) south of the Dalj settlement and one soil profile (P-10) near the Zmajevac settlement were excavated. The investigation included a detailed pedological analysis, a modal analysis of the heavy and light mineral fraction and a mineralogical analysis of bulk samples (the < 2 mm fraction) and the fraction < 2 μm. By comparing the obtained results with the criteria of the Croatian Soil Classification and the World Reference Base for Soil Resources, the soil profiles P-3 and P-6 can be defined as Chernozem on Loess or Hortic Calcic Chernozem (Epiloamic, Endosiltic, Aric, Humic). The systematic unit for profile P-10 was defined as Rendzina according to the Croatian Soil Classification or Calcic Chernozem (Siltic) according to the WRB. Based on the results of the pedological analysis of the soil profile horizons, a gradual degradation of the chernozem was observed as a result of anthropogenic influence, but also due to recent climate change. The degradation is particularly evident in the form of a reduction in organic matter and the relocation of carbonates from the surface to deeper zones. Due to the increasing degree of weathering caused by recent climate changes, some differences in the mineralogical composition of the studied soils were also observed. The progressive degradation of the chernozem due to the effects of recent weathering processes is indicated mainly by the presence of goethite in the fraction < 2 μm as a weathering product of iron minerals (magnetite, pyroxenes…). Although the parent material of all three profiles is loess sediments, the reason why the soil material of profile P-10 has not developed a chernic horizon is the constant contribution of aeolian material and a short period of exposure to pedogenetic processes.

## 1. INTRODUCTION

Globally, loess is a homogeneous rock concerning its sedimentological properties, geochemical and mineral composition. Geomorphologically, it is mostly found today on plateaus (mainly on river terraces) or on gentle slopes. The spatial distribution of palaeosols intercalated in the loess indicates a similar topography during the Pleistocene as it is in the present. Generally, the surface was covered by steppe during the cold periodes and by forest during the warm periods (BRONGER, 2003). Local differences in the mineralogical composition of loess/palaeosol sections in Eastern Croatia were established (GALOVIĆ & PEH, 2016). They were a consequence of the different source areas of aeolian material.

Loess/palaeosol successions, as a record of climatic changes, have been investigated in the area of eastern Croatia since the end of the 19$^{th}$ century (PILAR, 1875; GORJANOVIĆ-KRAMBERGER, 1911, 1922; ŠANDOR, 1912) and are still the focus of investigation (GALOVIĆ et al., 2009, 2011; GALOVIĆ, 2014, 2016; GALOVIĆ & PEH, 2016).

JENNY (1941) defined five factors influencing pedogenesis: climate, organisms, parent material, relief and time. Analyzing available factors and minorizing their variations could give us an insight into the length and intensity of warming periods during the Pleistocene.

The scope of this work is, through detailed pedological and mineralogical analyzes, to establish possible local differences in the parent materials of the recent leoss-derived soils. The soil profiles are located on a plain to avoid differences caused by relief. Since the organisms present in the soil depend on the prevailing climate, the type of mineralogical composition of palaeosol that developed during warming is a function of time and climate (GALOVIĆ & PEH, in press).

According to Dokuchaev, the Chernozems were defined as steppe soils. Their pedogenesis was mainly characterised by the soil-forming factors of the dry continental climate, steppe vegetation and loess as a carbonaceous parent material (DOKUCHAEV, 1948). It is generally assumed that his definition, although referring to Russian soils, can be generally applied to Eastern and Central Europe.

In publications dealing with the problem of chernozem in Central Europe, its existence in Croatia is usually not mentioned (e.g., NEJGEBAUER, 1983; ECKMEIER et al., 2007; ŁABAZ et al., 2018, 2022). However, according to the soil classification formerly used in Croatia (originally the Classification of Soils of Yugoslavia, ŠKORIĆ et al., 1973, 1985) and the Croatian Soil Classification (CSC) (HUSNJAK, 2014), chernozem occurs in Croatia. The reason for the rare mention is probably that it is a relatively small area and has not been reserched and worked on for more than 60 years. In fact, the chernozem soil type occupies an area of only about 50,000 ha in Croatia (BOGUNOVIĆ et al., 1988; HUSNJAK, 2014). About 95% of the Chernozem soil has long been used as arable land in intensive agriculture and the remaining 5% is mainly anthropogenic grassland. The presence of chernozem in Croatia according to the national soil classification is also mentioned by other authors but without detailed research (GRAČANIN, 1951; ŠKORIĆ, 1960; ĆIRIĆ, 1965; ŠKORIĆ et al., 1977; BOGUNOVIĆ et al. 1998; ŠPOLJAR et al. 2001; BAŠIĆ, 2013; PERNAR, 2017).

It is common knowledge that the most important type of soil for food production in the world is chernozem. Therefore, it has been used in agriculture for a long time both globally and in Croatia (NEJGEBAUER, 1951; ŠKORIĆ, 1960; ŠKORIĆ et al., 1977; ALTERMANN et al., 2005; HUSNJAK, 2014; POZNAIK, 2019). Chernozem is considered one of the most fertile agricultural soils in the world. Consequently, chernozem is one of the most useful soils for agriculture and contributes to high agricultural yields.

Furthermore, in Croatia, where chernozem occurs, there are other soils that are very similar to chernozem, but which, according to CSC, do not meet the criterion of the depth of the A horizon (> 40 cm) to be classified as chernozem. Therefore, we would like to point out the existence of chernozem and chernozem-like soils in Croatia and the basic physical, chemical and mineral properties of these soils.

The results of this research will be the starting point for the correlation of modern pedogenesis (as a reflection of modern climate) with palaeopedogenesis (as a reflection of palaeoclimate). This will significantly facilitate the palaeoclimatic reconstruction of the warming periods recorded in the loess/palaeosol sequences during the Pleistocene at the local level.

### 1.1. Geological and geographical setting

Quaternary sediments are widespread in eastern Croatia and include alluvial, marsh and lake sediments, often covered by aeolian silt-rich loess sediments formed during the Pleistocene cold periods (Fig. 1a). An area with extensive loess thickness is located in the "loess plateau" in eastern Croatia. The Dalj profiles are located in the east, and the Zmajevac profile is located in the northeast of eastern Croatia. In the Baranja area, neotectonic movements uplifted the Bansko brdo, exposing loess-palaeosol sequences up to 30 m thick on the south-eastern edge of the hill (HEĆIMOVIĆ, 1991), where a microlocation of the Zmajevac profile is located (Fig. 1b). In the Croatian lowlands, aeolian sediments were deposited in lakes, ponds and shallow swamps during the Quaternary (BAČANI et al., 1999) until they were infilled. Then aeolian sedimentation continued and loess sessu stricto formed (BAČANI et al., 1999; GALOVIĆ et al., 2009). Some of these sediments were eroded by the Danube, Drava and Sava rivers and/or redeposited further downstream as alluvial sediments. Similar deposits have been reported and studied from the Abony section in Hungary (FRECHEN & PÉCSI, 2004). The microlocation of the Dalj profiles is in the Croatian lowlands near the bank of the Danube.

The climate of the region is continental and the annual rainfall is about 700 mm (JELIĆ & KALOGJERA, 2002). Three analyzed profiles are located in the area, which is characterized by a temperate continental climate with dry summers (PEEL et al., 2007). The area is known as the driest part of the country and differs from central and western Croatia which is influenced by continental climate (PERČEC et al., 2023). NW and NE winds prevail at both sites. The mean annual wind speed is 4.6-4.8 $ms^{-1}$ (CROATIAN METEOROLOGICAL AND HYDROLOGICAL SERVICE, 2023).

The mineralogical and geochemical composition of the loess as parent material is consistent at both sites and has been described in detail in numerous publications (GALOVIĆ, 2014, 2016; GALOVIĆ & PEH, 2016).

#### Dalj

The bedrock of the Daljska planina (Dalj hill) consists of Miocene conglomerates, limestones and sandstones covered by clayey and sandy sediments from the Lower Pliocene (VELIĆ et al., 1985). At the beginning of the Quaternary, the subsidence rate increased, resulting in intense sedimentation. Tectonic activity during the Middle and Upper Pleistocene caused an uplift of the Erdut hill, while the northern and southern slopes underwent relative subsidence (VELIĆ et al., 1985; TRIFUNOVIĆ, 1985; BAČANI et al., 1999).

The Dalj profiles (P-3: 45°26'14" N. Lat., 18°58'10" E. Long. and P-6 45°26'46" N. Lat., 18°56'34" E. Long.) are located about 5 km south-southwest of the center of Dalj village (Fig. 1b). Profile P-3 is at an altitude of 88.7 and profile P-6 at an altitude of 88.3 m above sea level.

#### Zmajevac

The Bansko brdo is an asymmetric horst, tectonically elongated in a NE-SW direction and has a height of about 243 m a.s.l. Tectonic activity led to synsedimentary effusion of basaltic andesite and deposition of volcanic breccias. The Miocene age of the andesites is confirmed by K-Ar dating (14.5±0.4 and 13.8±0.4 Ma at two sites) (PAMIĆ & PIKIJA, 1987). Loess is exposed over the volcaniclastic material. Recent tectonic uplift has formed a complex horst, the Bansko brdo. These neotectonic movements are still active (HEĆIMOVIĆ, 1991).

The studied profile is located about 2 km NE of the village of Zmajevac (45°48'49" N. Lat., 18°49'33" E. Long.), at an altitude of 124 m above sea level.

### 1.2. Pedological setting

In Croatia, chernozem occurs only in the extreme northeast (Fig. 2a, b), where it developed on carbonate loess with a high content of primary carbonates under conditions of a continental climate and grassland vegetation. It is assumed that the chernozem in Croatia was a relict soil formed when the continental climate was characterized by dry summers and colder winters compared to today's climate. Such conditions enabled the humification of organic matter and contributed to the formation of a deep and humus-rich A horizon (GRAČANIN, 1951; NEJGEBAUER, 1951; ŠKORIĆ, 1960).

The time of the formation of the chernozem in Central Europe, i.e. its absolute age, cannot yet be determined with certainty. A majority of authors of publications consider the early Holocene as the time of chernozem formation (ROESCHMANN et al., 1982; SCHEFFER & SCHACHTSCHABEL, 2002), i.e., they believe that the formation of the chernozem ended around 5.500 years BC. This is because during this period there were favourable climatic conditions for the formation of a deep Mollic A horizon. The climate conditions during this period were characterised by warm and humid springs, which allowed the growth of large amounts of biomass, i.e., dry and hot summers and cold winters, during which more intensive mineralisation of organic matter was prevented/limited. Although it occupies only a small



**Figure 1.** a) Location of the studied soil profiles in Eastern Croatia (Europe Relief Map, maps-for-free.com last accessed on June 21, 2022); b) Geological Map of Eastern Croatia (Croatian Geological Survey, 2009) with the positions of the soil profiles in Zmajevac – P-10 and in Dalj P-3 and P-6).

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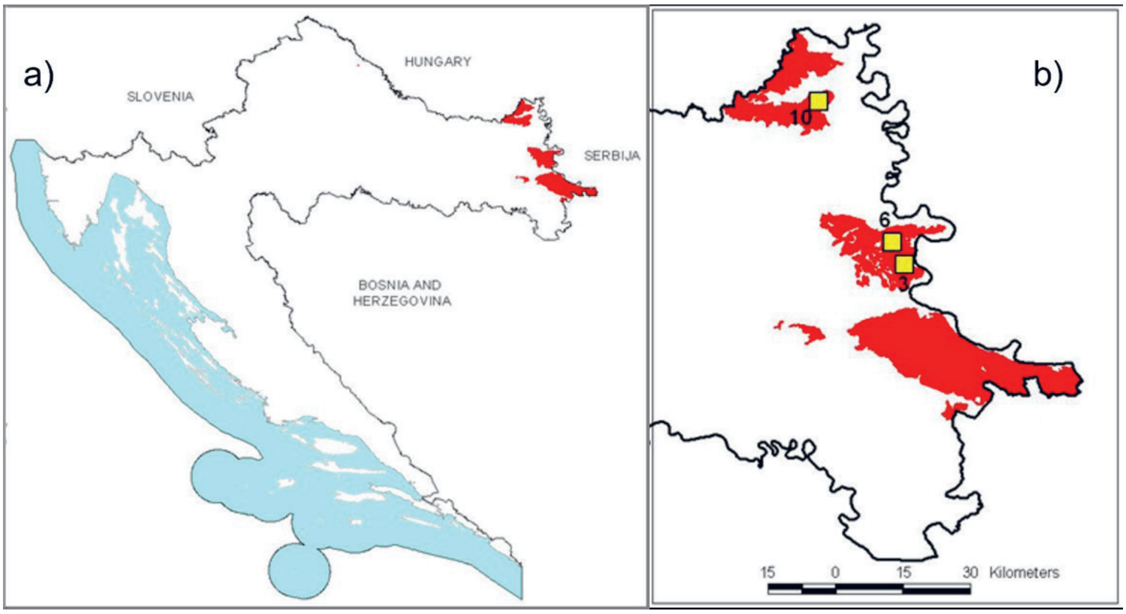


**Figure 2.** a) The area with a dominant presence of chernozem, with occasional occurrences of rendzina on the loess; b) Locations of soil profiles P-3, P-6 and P-10.

area (0.9 %), chernozem is an extremely important soil in Croatia. The composition of typical chernozem in Croatia is A-AC-C.

Confirmation of the existence of chernozems in Croatia is provided, for example, by work referring to the existence of chernozems in neighbouring Serbia, which Croatia borders (GAJIĆ et al., 2006; BOKHORST, et al., 2009; VIDOJEVIĆ et al., 2016; RADAKOVIĆ et al., 2019). In the area where chernozem is distributed in Croatia, there is soil similar to chernozem, characterized by the presence of a humus accumulative horizon < 40 cm deep, and formed under the same conditions as chernozem. According to the Croatian Soil Classification, such soil cannot be classified as chernozem due to the above criteria, but is classified as rendzina on loess, the composition of which is mostly A-C, more rarely A-AC-C. Since such soils can be classified as chernozems according to the World Reference Base for Soil Resources (IUSS WORKING GROUP WRB, 2022), they are treated as chernozem-like soils in this document. The area of this soil is not known, but it is estimated that it is not large.

## 2. METHODS

### 2.1. Pedological analyses

For the study of chernozem, two soil profiles were dug south of the Dalj settlement, and for the study of rendzina - chernozem-like soils, one soil profile was excavated near the Zmajevac settlement (Figs. 1b, 2b). For the pedological and mineralogical analyses, soil samples were taken in disturbed and undisturbed conditions according to the pedogenetic horizons. The pedological analyses were carried out in the laboratory of the Department of Soil Science, Faculty of Agriculture at the University of Zagreb. Various types of soil analyses were carried out according to standard methods. Soil samples were prepared according to HRN ISO 11464 (2009); determination of soil particle size distribution (mechanical composition), i.e. fine earth, was carried out using sieving and sedimentation methods according to HRN ISO 11277 (2011) with evaluation of texture classes according to FAO (2006); the stability of structural microaggregates was determined according to the Vageler method (JDPZ, 1971; ŠKORIĆ, 1982); the determination of the volume density of dry soil was carried out according to HRN ISO 11272 (2017); the determination of the density of solid particles was carried out according to HRN ISO 11508 (2004); determination of water retention capacity of soil, total pore content in soil and air capacity of soil was performed according to the Gračanin method (JDPZ, 1971; ŠKORIĆ, 1982); pH determination ($H_2O$ 1M KCl) was performed according to HRN ISO 10390 (2005); humus content was determined according to the Tjurin method (JDPZ, 1966); determination of carbonate content - volumetric method was performed according to HRN ISO 10693 (2004). The organic carbon content was calculated from the humus content using the Van Bemmelen conversion factor (humus content/1.724). The colour of the soil in the wet and dry state was determined according to the MUNSELL SOIL COLOR CHARTS (2013) and the structure according to FAO (2006).

### 2.2. Mineralogical analysis

#### 2.2.1. Modal analysis

To determine the qualitative and semi-quantitative mineral composition of heavy and light mineral associations, ten samples of pedgenetic horizons (four from the P-3 and four from the P-6 soil profiles in Dalj and two from the P-10 profile in Zmajevac) were examined.

After disaggregation in an ultrasonic bath and sieving to the size fraction 0.09-0.125 mm, the calcite was dissolved. This fraction was selected for analysis because it contains all the virtual mineral species in a ratio representative of the bulk sample. The heavy mineral fraction (HMF) was separated with sodium polytungstate (SPT) ($\rho = 2.8$ g cm$^{-3}$). The slides of the heavy and light mineral fraction (LMF) were examined with the polarising microscope AxioLab.A1 from Carl Zeiss. The qualitative and semi-quantitative composition of a sample was determined after 300-400 grains were identified and the percentage of each mineral was calculated. Since the specific gravity of muscovite is between 2.76 and 3.00 g cm$^{-3}$, it is predominantly a component of HMF, but some crystals remain in LMF. Therefore, the number of muscovite crystals counted as a component of LMF was multiplied by the percentages of LMF and added to the percentages of muscovite in HMF. Canada balsam was used as the embedding medium.

The weathering index (W.I.) was used to examine the degree of alteration of the analysed soil horizons. The W.I. was defined

**Figure 3.** Landscape with the corresponding soil profile P-3.

by BREWER (1976) and applied by FAIVRE et al. (2019) and GALOVIĆ & PEH (in press) as the ratio of the percentages of resistant and non-resistant minerals: W.I. = (Zrn + Tur + Rt + Ttn + St + Grt) / (Ep-Zo + Amp + Px + Ky). It is calculated to four decimal places on the basis of the percentage contents of the minerals analysed. A higher W.I. indicates significant or repeated weathering of the analysed grains due to long exposure to a warm and humid geochemical environment (pedogenesis) and/or resedimentation.

### 2.2.2. X-ray diffraction method

The mineral composition of the soil samples of the fractions < 2 mm and < 2μm was measured by X-ray powder diffraction (XRD) with a PANalytical X' Pert PRO diffractometer, equipped with a Cu-tube, graphite monochromator and Pixel detector. Samples of the fraction <2 mm (bulk samples) were dried, crushed and homogenised in an agate mortar to the powder fraction. The fraction <2 μm was separated from the insoluble residue by centrifugation after dissolving the carbonates with a 1 M NaOAc solution buffered to pH 5 with HOAc and removing the organic matter with $H_2O_2$. The time and number of revolutions were determined according to Stocks law. XRD patterns of the clay fraction were obtained on oriented mounts after different treatments: (a) air drying, (b) ethylene glycol solvation of air dried samples, (c) $K^+$ saturation with 4M KCl solution, (d) $Mg^{2+}$ saturation with 4M $MgCl_2$ solution, (e) DMSO solvation of K-saturated samples, (f) ethylene glycol solvation of K-saturated samples, (g) solvation of Mg-saturated samples with ethylene glycol, (h) solvation of Mg-saturated samples with glycerol, (i) heating to 400°C for 1h, (j) heating of the saturated samples to 350°C and (k) heating to 550°C for 1h. According to BROWN (1961), BRINDLAY & BROWN (1980), MOORE & RAYNOLDS (1997), VELDE & MEUNIER (2008) and HARIS & WHITE (2008), these treatments are a very efficient means of identifying and distinguishing clay minerals. Semi-quantitative estimates of minerals detected in bulk samples were determined by Rietveld refinement using *PANalytical HighScore Plus* software linked to the ICCD mineralogical database. Semi-quantitative estimates of clay minerals in the fraction <2μm were determined from the relative intensities of characteristic X-ray reflections using the method of JOHNS et al. (1954), where the number of plus signs represents their relative abundance in the sample. The abbreviations for the mineral names are used according to WHITNEY & EVANS (2010).

## 3. RESULTS

### 3.1. Soil properties

According to the Croatian Soil Classification (HUSNJAK, 2014), the physico-chemical and mineralogical properties of chernozem are presented on the basis of two soil profiles and those of rendzina on loess (chernozem-like soil) based on one soil profile.

**Soil profile P-3** is characterised by the structure of the Ap-A-AC-C profile, where the surface part of the A horizon is anthropogenic down to a depth of about 42 cm and represents the arable horizon. The depth of the A horizon is about 65 cm (Fig. 3).

**Table 1.** Mechanical composition of soil and stability of macroaggregates.

| Profile number | Horizon | Depth (cm) | Content of mechanical particles (%) in Na-pyrophosphate (mm) | | | | | [1]Texture | Stability of microaggregates | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | 2.0-0.2 | 0.2-0.063 | 0.063-0.02 | 0.02-0.002 | <0.002 | | [2]Ss | Assessment |
| P-3 | Ap | 0-42 | 0.2 | 3.1 | 37.9 | 31.7 | 27.1 | PrGl | 62.0 | fairly stable |
| | A | 42-65 | 0.3 | 2.8 | 36.9 | 32.2 | 27.8 | PrGl | 55.3 | fairly stable |
| | AC | 65-110 | 0.6 | 2.9 | 38.6 | 33.4 | 24.5 | Prl | | - |
| | C | 110-150 | 0.7 | 9.1 | 42.1 | 32.0 | 16.1 | Prl | | - |
| P-6 | Ap | 0-40 | 0.2 | 2.4 | 35.8 | 34.3 | 27.3 | PrGl | 56.9 | fairly stable |
| | A | 40-72 | 0.2 | 4.8 | 33.7 | 33.7 | 27.6 | PrGl | 51.4 | fairly stable |
| | AC | 72-106 | 0.3 | 3.5 | 35.6 | 36.4 | 24.2 | Prl | | - |
| | C | 106-140 | 0.5 | 5.1 | 40.8 | 36.6 | 17.0 | Prl | | - |
| P-10 | A | 0-30 | 0.5 | 6.4 | 51.7 | 28.4 | 13.0 | Prl | 64.1 | fairly stable |
| | C | 30-100 | 0.3 | 7.5 | 51.6 | 28.3 | 12.3 | Prl | 28.9 | slightly stable |

[1]Prl - silty loam; PrGl - silty clay loam; [2]Ss - stability index

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**Table 2.** Structure and colour of the soil.

| Profile number | Horizon | Depth (cm) | Structure | Colur | |
|---|---|---|---|---|---|
| | | | | In the wet state | In the dry state |
| P-3 | Ap | 0-42 | granular | 10YR 3/2 | 10YR 5/2 |
| | A | 42-65 | granular | 10YR 3/2 | 10YR 5/2 |
| | AC | 65-110 | granular to crumbly | 10YR 4/2 | 10YR 6/3 |
| | C | 110-150 | undeveloped | 10YR 6/3-10YR 8/3 | 10YR 7/3-10YR 8/4 |
| P-6 | Ap | 0-40 | granular | 7.5YR 3/1 | 10YR 4/2 |
| | A | 40-72 | granular | 10YR 3/2 | 10YR 6/2 |
| | AC | 72-106 | granular to crumbly | 2,5YR 4/3 | 10YR 6/2 |
| | C | 106-140 | undeveloped | 10YR 8/4-7,5YR 5/3 | 10YR 8/3-7,5YR 6/4 |
| P-10 | A | 0-30 | granular | 10YR 3/2 | 2.5Y 5/2 |
| | C | 30-100 | undeveloped | 10YR 6/3 | 2.5Y 7/4 |

**Table 3.** Basic physical properties of soil.

| Profile number | Depth (cm) | $^1\rho_b$ g/cm$^3$ | $^2\rho_s$ g/cm$^3$ | $^3$P | $^4$SWC | $^5$AC |
|---|---|---|---|---|---|---|
| | | | | % vol | | |
| P-3 | 0-42 | 1.44 | 2.62 | 45.0 | 36.6 | 8.4 |
| | 42-65 | 1.32 | 2.68 | 50.9 | 39.8 | 11.1 |
| | 65-110 | 1.25 | 2.69 | 53.6 | 42.2 | 11.4 |
| | 110-150 | 1.19 | 2.65 | 55.2 | 37.7 | 17.5 |
| P-6 | 0-40 | 1.27 | 2.66 | 52.3 | 38.6 | 13.7 |
| | 40-72 | 1.21 | 2.68 | 54.9 | 41.7 | 13.3 |
| | 72-106 | 1.23 | 2.73 | 55.0 | 40.9 | 14.1 |
| | 106-140 | 1.22 | 2.72 | 55.2 | 40.1 | 15.1 |
| P-10 | 0-30 | 1.25 | 2.65 | 53.1 | 38.0 | 15.1 |
| | 30-100 | 1.21 | 2.70 | 55.2 | 43.4 | 11.8 |

$\rho_b$ - volume density; $^2\rho_s$ – soil particle density; $^3$P - total porosity; $^4$SWC - soil water capacity; $^5$AC - air capacity

The soil texture in the Ap and A horizons is silty clay loam, i.e. at the boundary between silty clay and silty clay loam. Deeper in the soil profile it is silty loam. The clay particle content determined in the C horizon -parent substrate (loess) is significantly lower than in the upper horizons, where the reduction is about 40% (Table 1). The stability of the structural microaggregates is fairly stable in the arable soil as well as in the lower part of the A horizon. The soil structure in the Ap and A horizons is granular and the colour is very dark grayish brown (Table 2).

As a result of the anthropogenic influence on the soil by trampling, porosity is lowest in the surface Ap horizon and then increases with depth (Table 3). The water capacity of the soil changes slightly with depth down to the C horizon, where it is the lowest. The air capacity of the soil is broadly similar in trend to porosity, being lowest in the Ap horizon and highest in the C horizon - the parent substrate.

This soil is carbonate-rich throughout the depth of the profile, although the carbonate content in the Ap horizon is significantly lower compared to the other horizons. The content in the Ap horizon is only 8% of the total carbonate content in the parent substrate (Table 4). In the zone between 50 and 100 cm, the existence of secondary carbonates in the form of $CaCO_3$ concretions and pseudomycelia was observed. Accordingly, the soil reaction is alkaline, with the soil pH being lowest in the Ap horizon and increasing with depth. The organic carbon content is highest in the Ap horizon, followed by horizons A and AC (Table 4).

**The soil profile P-6** is also characterized by the horizon sequence Ap-A-AC-C, whereby the surface part of the A horizon is also anthropogenic to a depth of about 40 cm (Fig. 4).

The soil texture is very similar to that of soil profile P-3. The Ap and A horizons are silty clay loam, i.e. at the boundary between silty clay and silty clay loam. In the deeper horizons, the texture is silty loam. It is also characteristic of this profile that the content of clay particles in the parent substrate of the C horizon is significantly lower than in the upper horizons, where it amounts to about 35% (Table 1). The structural microaggregates are fairly stable in the arable Ap horizon as well as in the remaining part of the A horizon. The soil structure in the Ap and A horizons is

**Figure 4.** Landscape with the corresponding soil profile P-6.

**Table 4.** Basic chemical properties of the soil.

| Profile number | Depth (cm) | pH $H_2O$ | pH KCl | $CaCO_3$ (%) | Humus (%) | SOC[1] (%) |
|---|---|---|---|---|---|---|
| P-3 | 0-42 | 8.31 | 7.40 | 2.12 | 2.51 | 1.46 |
| | 42-65 | 8.41 | 7.77 | 19.52 | 1.81 | 1.05 |
| | 65-110 | 8.51 | 7.83 | 25.88 | 1.01 | 0.59 |
| | 110-150 | 8.60 | 7.99 | 26.31 | 0.18 | 0.10 |
| P-6 | 0-40 | 8.23 | 7.44 | 2.97 | 2.86 | 1.66 |
| | 40-72 | 8.41 | 7.75 | 18.67 | 1.74 | 1.01 |
| | 72-106 | 8.34 | 7.88 | 26.13 | 1.07 | 0.62 |
| | 106-140 | 8.65 | 7.92 | 26.46 | 0.22 | 0.13 |
| P-10 | 0-30 | 7.91 | 7.56 | 15.60 | 2.12 | 1.23 |
| | 30-100 | 8.38 | 7.94 | 20.40 | 0.38 | 0.22 |

[1]Soil organic carbon

granular, and the color is dark brown in the Ap horizon and brown in the rest of the A horizon (Table 2).

The porosity of the soil barely increases with depth. As a result of anthropogenic compaction, it is lowest in the Ap horizon and then increases slightly with depth (Table 3). The water capacity of the soil also increases with depth down to the C horizon – the parent substrate, where it is slightly lower. The air capacity of the soil is lowest in the Ap horizon and increases with depth, being significantly higher in the C horizon compared to the upper horizons.

The soil is carbonate-rich throughout the depth of the profile, and the carbonate content in this profile is significantly lower in the Ap horizon compared to the other horizons (Table 4). Compared to the carbonate content in the parent substrate in the Ap horizon, it is only 11%. The soil acidity is correspondingly alkaline, with the soil pH being lowest in the Ap horizon and increasing with depth. Secondary carbonates were found in the zone from 50 to 100 cm in the form of pseudomycelia and $CaCO_3$ concretions in this soil profile. The organic carbon content is highest in the Ap horizon and decreases with depth (Table 4).

**Soil profile P-10** is characterized by the horizon profile A-C, with the depth of the A horizon being about 30 cm (soil is beneath natural vegetation).

The soil texture is silty loam in both the A and C horizons. Interestingly, there are no significant differences in the content of individual particles between the A and C horizons. The structural microaggregates are fairly stable in the A horizon, while in the C horizon the stability of the aggregates is low (Table 1). The soil structure in the A horizon is granular and in the C horizon it is not pronounced, while the soil colour is very dark grayish brown in the A horizon and pale brown in the C horizon (Table 2). Soil porosity and soil water capacity increase with depth, while soil air capacity and soil volume density decrease with depth (Table 3).

The soil is carbonate-rich throughout the depth of the profile, with lower carbonate content in the A horizon than in the C horizon. Accumulations of secondary carbonates in the form of pseudomycelia and nodules were found in the C horizon. In relation to the carbonate content of the parent substrate in the A horizon, it is about 68%. The soil acidity is correspondingly alkaline, with a slightly lower soil pH in the A horizon compared to the C horizon. The organic carbon content is significantly higher in the A horizon compared to its content in the parent substrate (Table 4).

**Figure 5.** Soil profile P-10.

## 3.2. Mineralogical characteristics

### 3.2.1. Modal composition

The LMF in almost all samples is > 95%, even if muscovite is reported as a heavy mineral (see section 2.2.1, Table 6 and Figs. 6 and 7). The dominant component in the light mineral fraction is quartz (74 - 83%), followed by feldspar (16 - 8%) and lithic fragments (12 - 5%). Comparing the proportions of weathered and fresh quartz grains, fresh grains account for 66% of all quartz in profiles P-3 and P-6 compared to 33% for weathered grains, while they are equally represented in profile P-10. Feldspars are almost exclusively represented by K-feldspars, mostly orthoclase, rarely microcline. Fresh feldspar grains predominate in the uppermost horizons of profiles P-3 and P-6 (Fig. 6a), while weathered feldspars predominate in all other horizons. The fresh grains are rich in elongated inclusions (Fig. 6b). In the rare cases where this was possible, they were determined to be idiomorphic zircon inclusions. Weathered feldspars are kaolinized and sometimes even sericitized and contain no inclusions (Fig. 6b). Plagioclase is represented by up to 1 % of Na-rich, equally weathered and fresh grains with a characteristic polysynthetic twinning. Lithic fragments are mostly represented by metamorphic rocks (undulose quartz, muscovite and chlorite (rarely biotite and feldspars)) and sporadically by quartzite or chert.

The contribution of HMF ranges from 1.85 to 5.54% and increases with depth in each profile. An exception is the uppermost horizon Ap in profile P-6. The highest proportion of HMF is found in profile P-10 (3.96–5.54%). Among the HMF, transparent heavy minerals predominate (69–87%). Opâque grains (5–12%) are rare in profile P-10 (5–8%) and are mainly represented by limonite. Chlorite (6–18%) is well rounded and often represents

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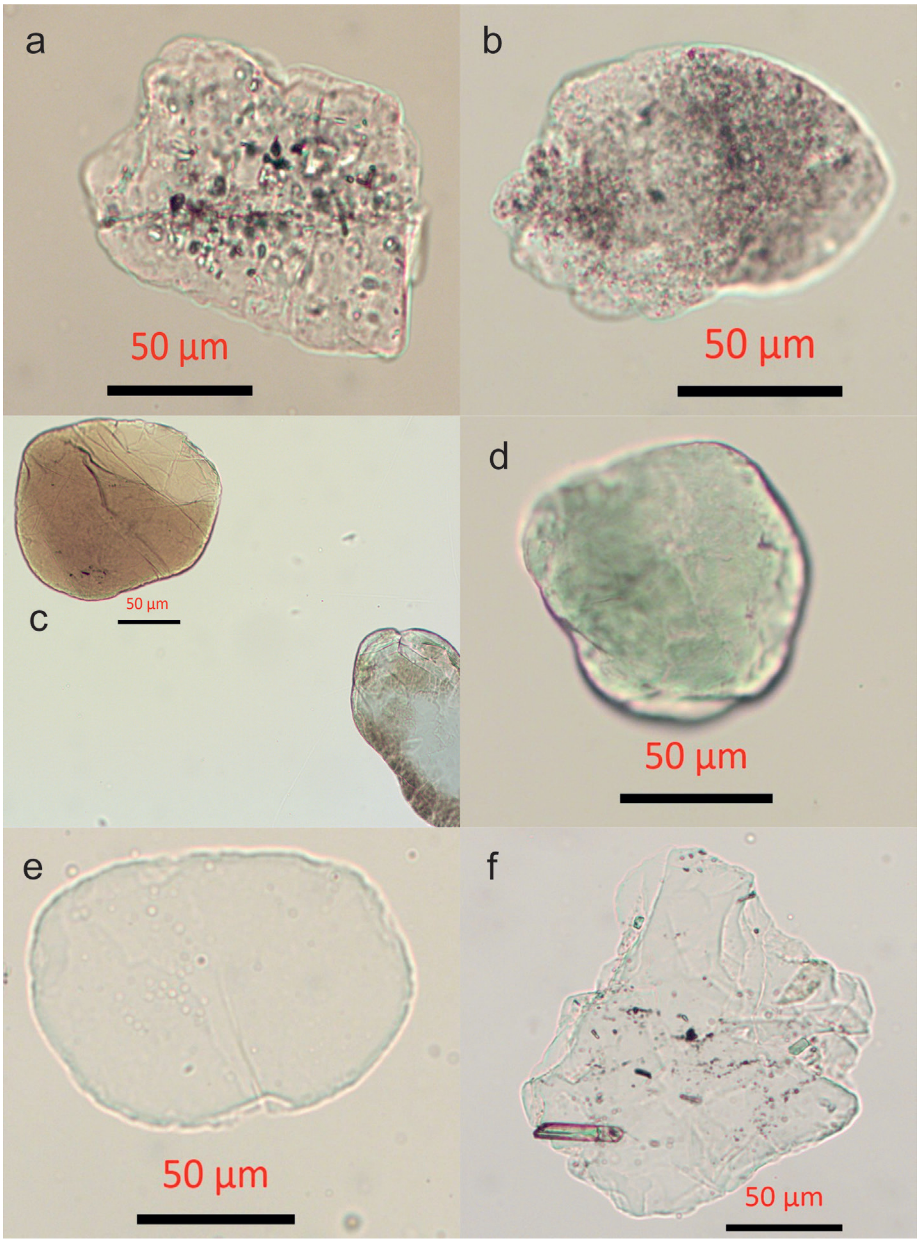


**Figure 6.** Photomicrographs of minerals from LMF and HMF of samples of the investigated profiles in polarized light: a) fresh K-feldspar rich in elongated inclusions – P3 65-110; b) weathered K-feldspar – P3 65-110; c) well-rounded fresh biotite (upper left) and chloritized biotite (lower right) – ZN A; d) well-rounded fresh chlorite; e) well-rounded muscovite – P3 110-150; f) muscovite with ragged rims containing elongated minerals with parallel extinction – ZN C.

**Table 5.** Modal composition of the light mineral fraction (values in %) and Weathering index (Legend in Table 6).

| Profile | Depth from the surface (cm) | Quartz | | Feldspar | | | | Lithic particles | | | [1]W.I. |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | K-feldspar | | Plagioclase | | | | | |
| | | fresh | weathered | fresh | weathered | fresh | weathered | quartzite | chert | other | |
| P-3 | 0-42 | 52 | 22 | 10 | 5 | + | 0 | 2 | 4 | 4 | 0.63 |
| | 42-65 | 46 | 26 | 6 | 9 | + | 0 | 1 | 2 | 9 | 0.71 |
| | 65-110 | 55 | 23 | 4 | 7 | + | 1 | 1 | 1 | 9 | 0.93 |
| | 110-150 | 62 | 21 | 5 | 7 | 0 | 0 | 2 | 1 | 2 | 1.00 |
| P-6 | 0-40 | 52 | 28 | 7 | 6 | 1 | 0 | 2 | 1 | 3 | 0.66 |
| | 40-72 | 53 | 27 | 5 | 7 | 0 | 1 | 2 | 1 | 4 | 0.63 |
| | 72-106 | 51 | 26 | 6 | 9 | 0 | 0 | 2 | 2 | 3 | 0.80 |
| | 106-140 | 55 | 28 | 4 | 5 | 1 | + | 1 | 1 | 6 | 0.69 |
| P-10 | 0-30 | 43 | 41 | 3 | 5 | 1 | 0 | 1 | 1 | 6 | 0.50 |
| | 30-100 | 39 | 37 | 4 | 9 | 0 | 1 | 0 | 1 | 9 | 0.70 |

[1]W.I. – Weathering index

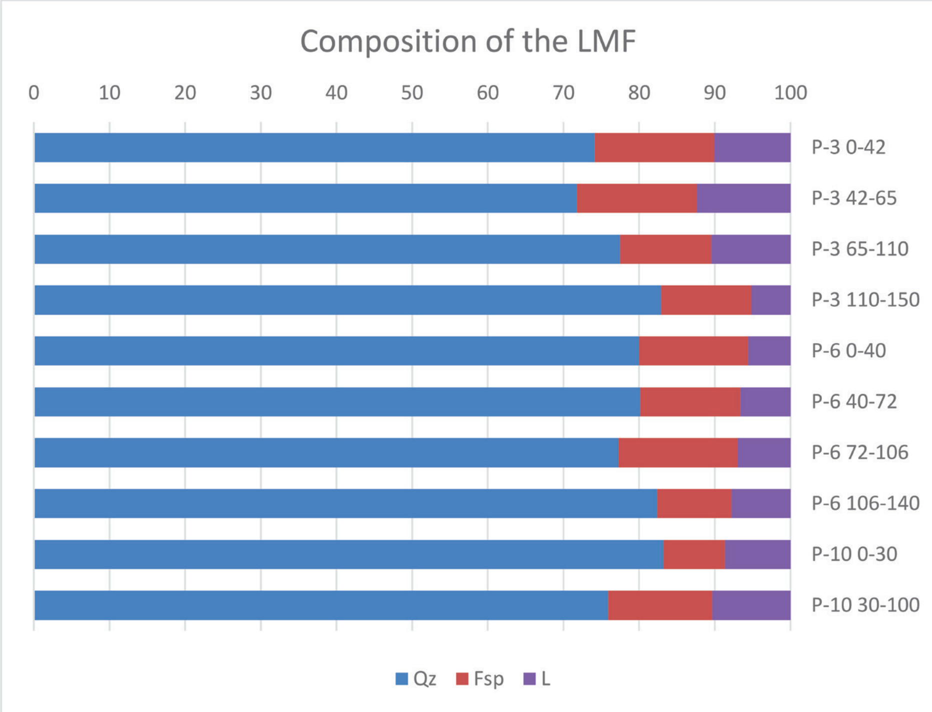


**Figure 7.** Modal composition of LMF (values in %). Legend in Figure 8.

**Table 6.** Modal composition of heavy and light mineral association.

| Profile | Depth from the surface (cm) | Composition of LMF 100% | | | HMF w % | Composition of HMF 100% | | | | Transparent heavy minerals 100% | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Qz | Fsp | L | | op | Chl | Bt | THM | Ms | Ep-Zo | Amp | Px | Grt | Ky | St | Tur | Zrn | Rt | Ttn | Chr | Ap |
| P-3 | 0-42 | 74 | 16 | 10 | 1.85 | 8 | 11 | 0 | 81 | 18 | 14 | 27 | 8 | 26 | 1 | 2 | + | + | 2 | 1 | 0 | + |
| | 42-65 | 72 | 16 | 12 | 2.65 | 11 | 10 | 1 | 79 | 28 | 17 | 16 | 8 | 19 | 0 | 3 | 1 | 3 | 3 | 1 | 0 | 2 |
| | 65-110 | 77 | 12 | 10 | 3.28 | 11 | 7 | 1 | 81 | 19 | 14 | 12 | 15 | 26 | 1 | 2 | 1 | 2 | 6 | 2 | 0 | 1 |
| | 110-150 | 83 | 12 | 5 | 4.85 | 5 | 8 | 1 | 87 | 11 | 14 | 18 | 12 | 34 | 0 | 2 | 2 | 2 | 2 | 2 | 0 | 0 |
| P-6 | 0-40 | 80 | 14 | 6 | 2.45 | 9 | 6 | 1 | 85 | 8 | 17 | 22 | 16 | 26 | 1 | 3 | 2 | 1 | 3 | 1 | + | 0 |
| | 40-72 | 80 | 13 | 7 | 1.91 | 8 | 12 | 1 | 79 | 17 | 12 | 25 | 12 | 24 | 2 | 2 | 3 | 0 | 1 | 2 | 0 | 1 |
| | 72-106 | 77 | 16 | 7 | 2.30 | 10 | 11 | 1 | 78 | 19 | 14 | 18 | 11 | 30 | 1 | 2 | + | 1 | 1 | 1 | 0 | 1 |
| | 106-140 | 82 | 10 | 8 | 2.64 | 12 | 15 | 0 | 73 | 15 | 15 | 24 | 11 | 25 | + | 2 | 2 | 1 | 3 | 1 | 0 | + |
| P-10 | 0-30 | 83 | 8 | 9 | 2.65 | 8 | 14 | 3 | 75 | 41 | 16 | 16 | 7 | 18 | + | 1 | 0 | 1 | + | + | 0 | + |
| | 30-100 | 76 | 14 | 10 | 3.28 | 5 | 18 | 7 | 69 | 65 | 5 | 12 | 3 | 12 | 0 | 1 | 1 | 0 | 0 | 0 | 0 | 0 |

Legend:
LMF = light mineral fraction, HMF = heavy mineral fraction, THM = transparent heavy minerals, Qz = quartz, Fsp = feldspar, l = transparent lithic particles, Op = opâque minerals, Chl = chlorite, Bt = biotite, Ms = muscovite, Ep-Zo = epidote–zoisite, Amp = amphibole, Px = pyroxene, Grt = garnet, Ky = kyanite, St = staurolite, Tur = tourmaline, Zrn = zircon, Rt = rutile, Ttn = titanite, Chr = chromite, Ap = apatite, + = minerals with occurrence < 0.5% (Symbology according to WARR (2021)).

chloritised biotite (Fig. 6c & d). Chlorite is most abundant in the P-10 profile (14–18%), which is also richest in biotite (3–7%). Biotite is present in all samples.

Muscovite is present in all samples, but its content varies considerably (8–65%). It is mostly represented by flakes and rarely by sericite. The muscovite flakes in profiles P-3 and P-6 are well-rounded (Fig. 6e), while the flakes from profile P-10 have ragged rims and contain many needle-like minerals with parallel extinction (Fig. 6f). Like other sheet minerals (biotite and chlorite), muscovite is most abundant in profile P-10 (41–65%). Primary chlorite crystals can be confirmed by the presence of primary chlorite in lithic fragments (metamorphic rocks). However, in many coloured leaflets, it was difficult to determine if it was still biotite or if it could be considered chlorite (chloritised biotite).

The most abundant transparent heavy minerals are resistant garnet grains (12–34%), followed by amphiboles (12–27%), the epidote-zoisite group (5–17%) and pyroxenes (3–16%). Garnet grains are often pink, sometimes colourless. Amphiboles are mostly represented by weathered dark green to olive green hornblende, sometimes by fresh brown to light olive brown amphiboles and sporadically by bluish purple glaucophane. Green hornblende grains are often chloritised. The Ap horizon of the P-3 profile is enriched in amphiboles. The epidote-zoisite group is represented by equal-sized, irregular, weathered grains. Epidote is generally yellow to greenish-yellow and shows weak pleochroism, while zoisite is colourless and shows a characteristic anomalous blue interference colour. The pyroxene group is dominated by clinopyroxene. Accompanying minerals are kyanite, staurolite, tourma-

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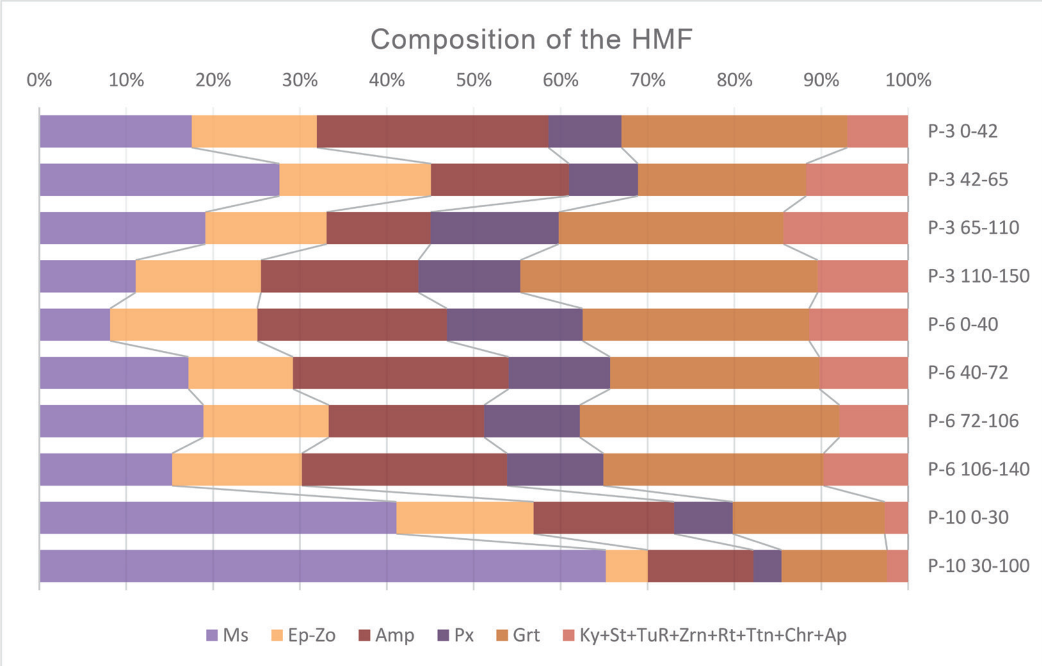


**Figure 8.** Modal composition of HMF (values in %).

Legend:

LMF = light mineral fraction, HMF = heavy mineral fraction, THM = transparent heavy minerals, Qz = quartz, Fsp = feldspar, l = transparent lithic particles, Op = opâque minerals, Chl = chlorite, Bt = biotite, Ms = muscovite, Ep-Zo = epidote–zoisite, Amp = amphibole, Px = pyroxene, Grt = garnet, Ky = kyanite, St = staurolite, Tur = tourmaline, Zrn = zircon, Rt = rutile, Ttn = titanite, Chr = chromite, Ap = apatite, + = minerals with occurrence < 0.5% (Symbology according to WARR (2021)).

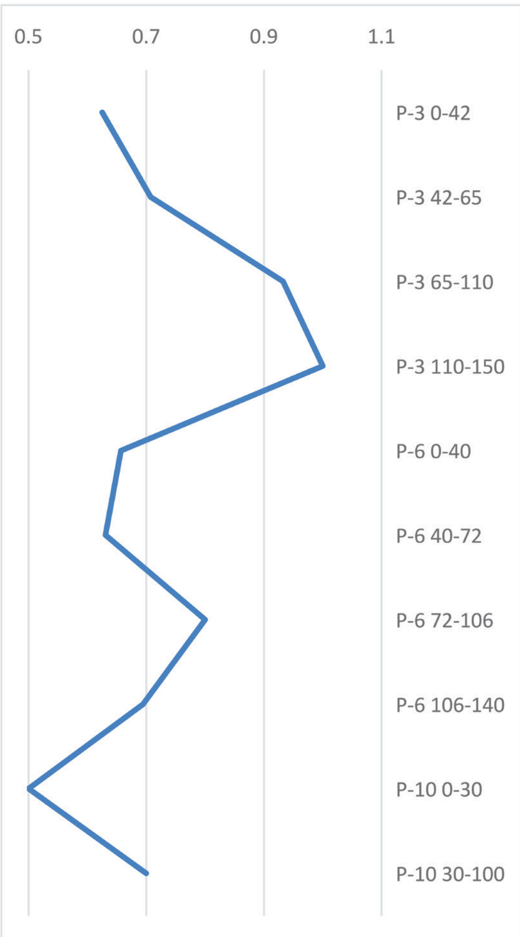


**Figure 9.** Distribution of W.I. (W.I. = (Zrn + Tur + Rt + Ttn + St + Grt) / (Ep-Zo + Amp + Px + Ky) in investigated soil profiles. (Legend in Table 6).

line, zircon, rutile and titanite. The results of the modal analyses are presented in Tables 5 and 6 and in Figs. 6, 7 and 8.

The uppermost Ap horizons of Dalj profiles P-3 and P-6 are characterised by sporadic spherical grains of devitrified volcanic glass in the LMF (Fig. 7) and weathered carbonate grains in the HMF (Fig. 8), in addition to a higher content of fresh quartz and feldspar grains. Since the specific gravity of calcite is 2.71 g $cm^{-3}$, dolomite is 2.85 ± 0.01 g $cm^{-3}$ and the applied heavy liquid is 2.8 g $cm^{-3}$, they are determined as dolomite grains.

The most important morphological feature of the analysed grains is that they are highly spherical, rounded, hypidiomorphic to allotriomorphic grains. This is most noticeable in the rounded habit of typical (hyp-)idiomorphic crystals such as tourmaline and zircon (Fig. 8).

The distribution of W.I. along the investigated soil profiles (Table 5; Fig. 9) shows a clear increase in W.I. along profiles P-3 and P-10 and an increasing trend along profile P-6.

### 3.2.2. Semiquantitative X-Ray Diffraction Analysis

The investigated XRD patterns of randomly oriented powder samples from profiles P-3 and P-6 show that quartz, micaceous minerals (muscovite/illite) and carbonates ($CaCO_3$) predominate (Table 7). Their content is largely uniform across the profile, with the exception of carbonates, which decrease sharply in the Ap horizon, while their content increases deeper in the profile. Plagioclase, potassium feldspar, dolomite and chlorite are present in lower amounts. Negligible contents of amphiboles are present in all samples. Unlike the profile of Dalj, samples from profile P-10 contain large amounts of quartz, carbonate minerals, micaceous minerals, and plagioclase. Potassium feldspars, amphiboles and other phyllosilicates are present in lower amounts. Compared with the profiles in Dalj this horizon contains a slightly larger amount of dolomite and amphiboles.

**Table 7.** Semiquantitative mineral composition of the fractions < 2 mm and < 2 µm.

| Profile | | Mineral composition of fraction < 2 mm | | | | | | | | Clay minerals in fraction < 2 µm | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Qtz | Cal | Dol | Plg | K-Fs | Amp | Ms/Il | Phyl + Gt* | Chl | Il | Vr | Sm | LCV/ HCS | $Kln_D$ | MLM | Gt |
| P-3 | Ap | 32 | 3 | 4 | 9 | 22 | 1 | 22 | 6 | ++ | +++ | + | | | + | | + |
| | A | 28 | 18 | 4 | 18 | 7 | ~ | 21 | 4 | ++ | +++ | + | | | + | | + |
| | AC | 26 | 25 | 7 | 12 | 5 | ~ | 20 | 5 | ++ | +++ | ++ | | | + | | + |
| | C | 26 | 26 | 8 | 13 | 8 | ~ | 14 | 5 | ++ | +++ | ++ | | + | + | | + |
| P-6 | Ap | 40 | 2 | 4 | 14 | 10 | 1 | 22 | 7 | ++ | +++ | + | | | + | | + |
| | A | 26 | 19 | 7 | 12 | 8 | 1 | 24 | 2 | ++ | +++ | + | | | | | + |
| | AC | 27 | 26 | 8 | 11 | 3 | 1 | 21 | 3 | ++ | +++ | ++ | | | | | + |
| | C | 27 | 26 | 10 | 12 | 3 | ~ | 20 | 2 | ++ | +++ | ++ | | + | | + | + |
| P-10 | A | 30 | 12 | 15 | 12 | 11 | 2 | 12 | 6 | ++ | +++ | ~ | | | + | | ~ |
| | C | 29 | 18 | 11 | 14 | 6 | 2 | 13 | 6 | ++ | +++ | | + | | + | | ~ |

Legend:
Qtz-quartz, Cal-calcite, Dol-dolomite, K-Fs-potassium feldspar, Plg-plagioclase, Amp-amphibole, Ms/Il-muscovite or illite (micaceous minerals), Il – illite, Chl-chlorite, Vr-vermiculite, Sm-smectite, Gt-goethite, Phyl – phyllosilicates, $Kln_D$ – well crystallized kaolinite (which reacted with DMSO), MLM- mixed layer minerals which could be chlorite–vermiculite ore chlorite–smectite, LCV/HCV – low charge vermiculite/high charge smectite, ~ - in trace, + - relative abundance of clay minerals within horizons based on X-ray diffraction (no quantitative value is assigned to +), * - based on the analysis of fraction < 2 µm.

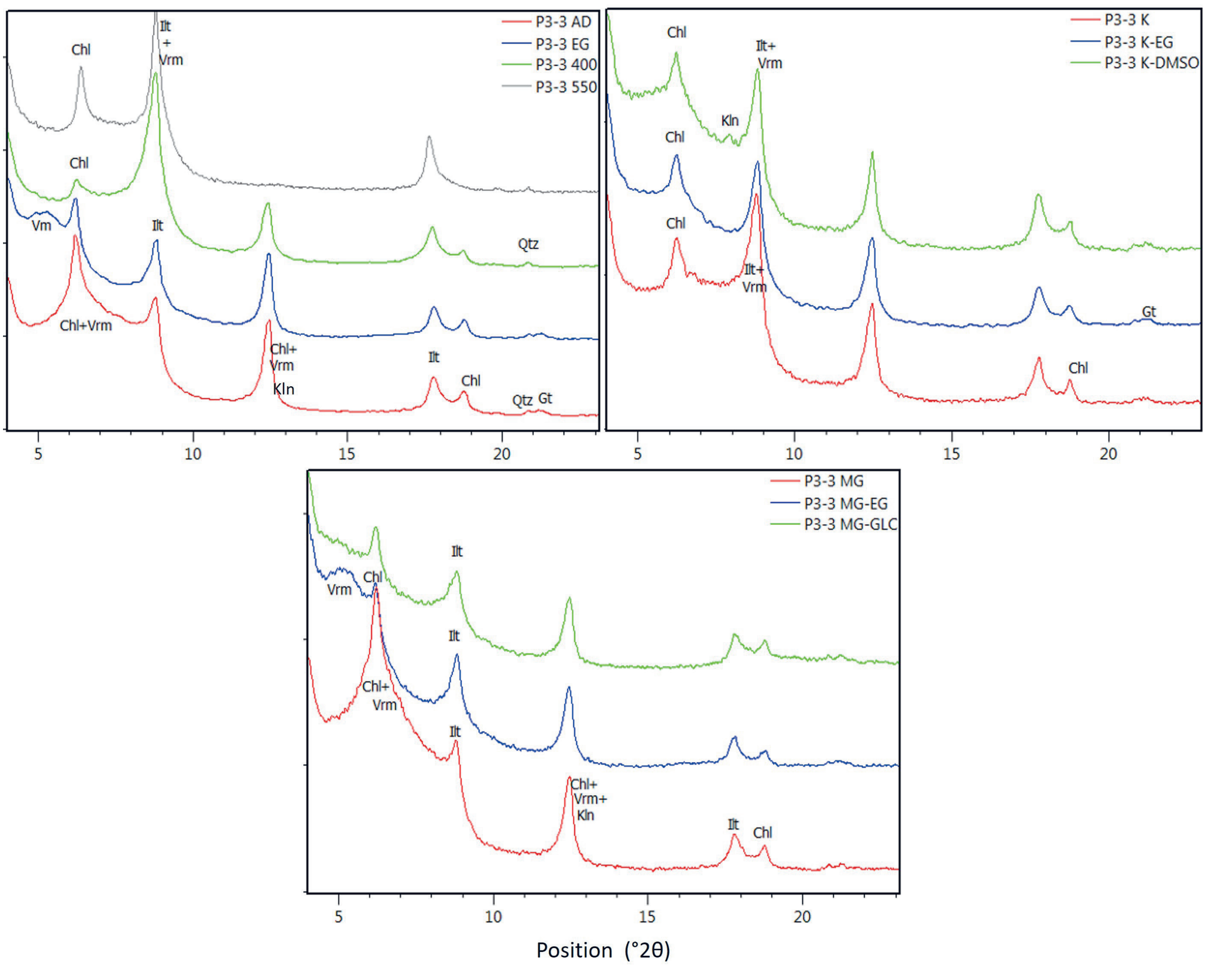


**Figure 10.** X-ray diffraction patterns of the clay fraction (< 2µm) from P-3 profile – horizon AC. AD – air dried, EG – ethylene glycol solvation, MG – saturated with Mg2+, K- saturated with K+, GLC – solvated with glycerole, DMSO – solvated with dimethyl sulfoxide, 400 – heated to 400°C, 550 – heated to 550°C. (Legend in Table 7).

Analysis of the fraction < 2µm determined that the most abundant clay minerals in all profiles are illite and chlorite. Their basal reflections at about 14 Å for chlorite and at 10 Å for illite are clearly visible after each treatment. Their content is constant over the entire length of the profiles. Except in the clay fraction, the clearly visible peak at 4.7 Å of bulk samples indicates the presence of chlorite in the silt fraction. In addition, the presence of expandable vermiculite was noted in all horizons, except in horizon C from profile P-10 were smectite is present. The content of vermiculite increases with depth, so it is most abundant in the

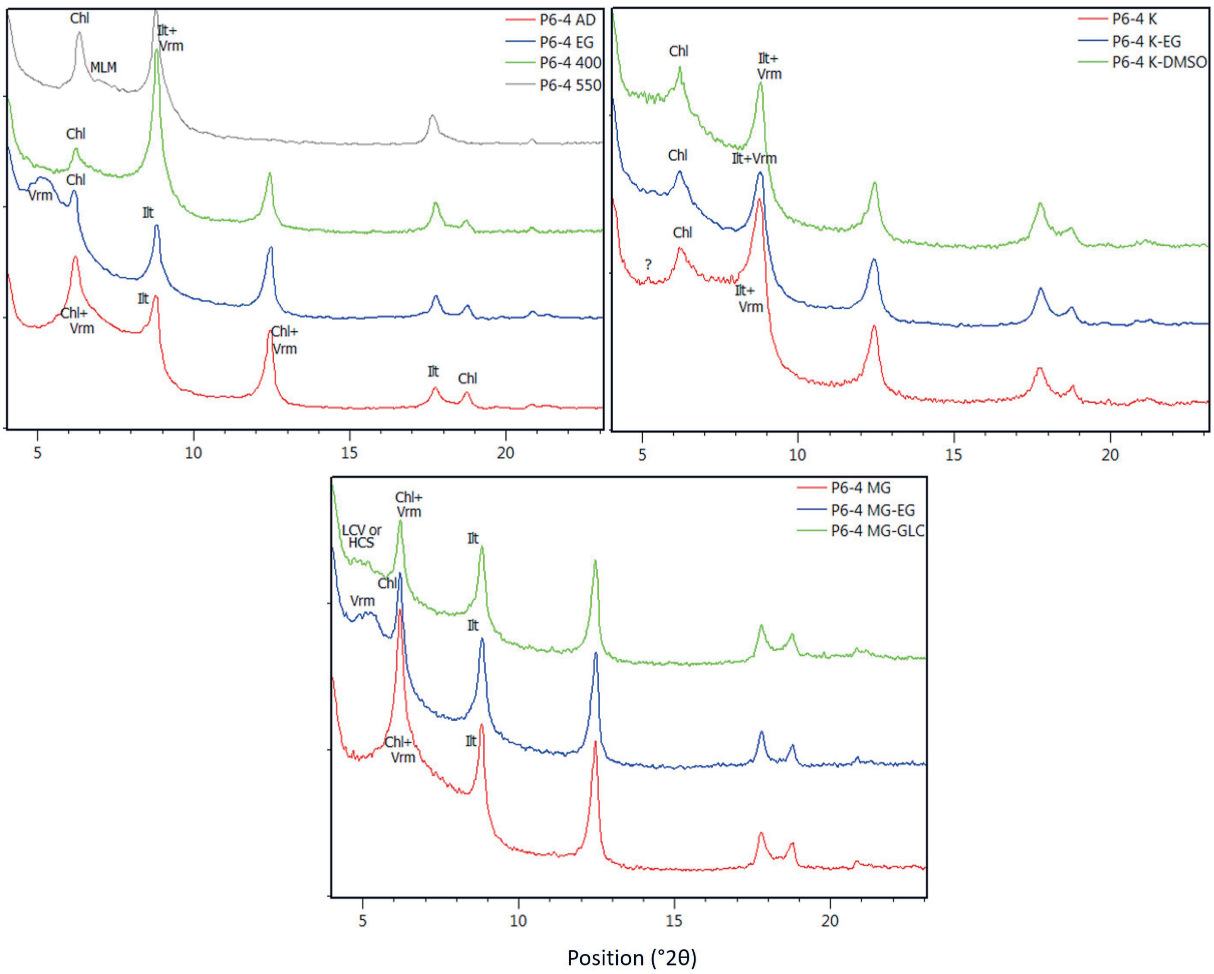


**Figure 11.** X-ray diffraction patterns of the clay fraction (< 2μm) from P-6 profile – horizon C.
AD – air dried, EG – ethylene glycol solvation, MG – saturated with Mg2+, K- saturated with K+, GLC – solvated with glycerole, DMSO – solvated with dimethyl sulfoxide, 400 – heated to 400°C, 550 – heated to 550°C. (Legend in Table 7).

horizons AC and C in profiles P-3 and P-6. Evidence for the presence of expandable vermiculite is the shift of the 14 Å reflections to the 17 Å position after glycolisation of air-dried and Mg-saturated samples, but the same is not observed in K-saturated samples (MOORE & RAYNOLDS, 1997). Furthermore, there is a clear sign of the presence of goethite in horizons from profiles P-3 and P-6, but only in the clay fraction. In profile P-10 the presence of goethite is in trace amounts. Its diffraction reflection is visible at 4.17 Å in all samples but disappeared after heating above 350°C (BRINDLAY & BROWN, 1980; MOORE & RAYNOLDS, 1997).

A minor amount of well-crystallised kaolinite (kaolinite that has reacted with DMSO) is found in all samples from the P-3 profile and in the A horizon of the P-6 profile (Fig. 10). The reflex at 7 Å of the samples saturated with DMSO may indicate pedogenetically formed kaolinite but also the chlorite and vermiculite. However, the clear and sharp peak instead of the small and very broad peak indicates that there is probably no or a very low presence of poorly crystallized kaolinite.

There are also some differences between the two analyzed soil profiles. In horizon C of the profile P-3, the presence of low charge vermiculite (LCV) or high charge smectite (HCV) was detected. Namely, after treatment of the Mg-saturated samples with glycerol, the 14 Å reflections were found to expand to 17–18 Å, but this effect was not visible in the reflections after ethylene glycol solvation of the K-saturated samples. It means that a clay mineral is present in the samples that has a characteristic of both vermiculite and smectite (MOORE & RAYNOLDS, 1997; TERHORST et al., 2012, DURN et al., 1999).

However, in a sample from horizon C in P-6 profile, the presence of mixed-layer clay minerals was detected on the basis of the occurrence of reflections at 12-13 Å after heating to 550°C, and it is most likely that this is some kind of interstratification between chlorite and vermiculite (Fig. 11; BRINDLEY & BROWN, 1980; MOORE & RAYNOLDS, 1997).

## 4. DISCUSSION

According to the criteria of the Croatian Soil Classification (HUSNJAK, 2014), the systematic unit for profiles P-3 and P-6 can be defined as chernozem on loess, carbonate, medium deep, unglazed, and anthropogenic. According to the criteria of the World Reference Base for soil resources (IUSS WORKING GROUP WRB, 2022) and based on the identified soil properties, the presence of a chernic horizon and a calcic horizon was determined in both profiles, based on which the pedosystematic unit can be defined as Hortic Calcic Chernozem (Epiloamic, Endosiltic, Aric, Humic).

The systematic unit for profile P-10 can be defined as Rendzina on loess, carbonate, medium deep, according to the above-mentioned soil classification of Croatia. As chernic and calcic horizons were also detected in this profile, the pedosystematic unit can be defined as Calcic Chernozem (Siltic) according to the WRB.

Research findings indicate a change or degradation of chernozem. Although it was established as early as 1960 that the chernozem in Croatia is gradually degrading, i.e. turning brown, due to higher temperatures and greater precipitation compared to the period of its formation (ŠKORIĆ, 1960), part of the chernozem is still carbonised. The research results indicate that the signs of degradation in the chernozem are mainly in the form of reduced carbonate content in the surface zone, which is result of centuries of continuous movement of carbonates from the surface zone to the subsurface zone.

It is assumed that the chernozem had > 4% humus in the A horizon at the time of its complete formation. However, due to climate change and human influence, especially during the last century, the humus content has decreased drastically. The significant influence of human activities is highlighted by DOKUCHAEV (1948), who believes that the rapid and intensive cultivation of chernozem began as early as the 17$^{th}$ century. The degradation of chernozem in Croatia is noted by ŠKORIĆ (1960), who found a humus content of only 2-3.5%, and NEJGEBAUER (1951), who found 2-4% humus in chernozem in the northern part of neighbouring Serbia (Vojvodina). Other authors also confirm the degradation of chernozem, for example KRUPENYKOV (2008); ŁABAZ et al. (2019), and KRAVICHENKO et al. (2012).

The significantly higher content of clay particles in the Ap, A, and AC horizons in both chernozem soil profiles compared to the content of clay particles in the C horizon, parent substrate, indicates the current processes of transformation of mineral matter with the formation of secondary clay minerals. At the same time, as already mentioned, the significantly lower content of carbonates in the Ap horizon compared to the other horizons and especially compared to the parent substrate indicates the current processes of carbonates displacement (leaching, migration). This means that carbonates are leached from the surface into the deeper horizons due to the processes of desilication and acidification of the soil. These processes can be a consequence of heavy rainfall, but also of agricultural activities, as is the case with the soils of profiles P-3 and P-6. The content of organic carbon indicates the presence of long-term processes of mineralization of organic matter in the soil in connection with the processes of humification, which also confirms the gradual degradation of the soil from the aspect of this property.

Based on the previously published analysis of the modal composition of loess as the parent material of the investigated recent soil profiles in the area investigated in this paper (GALOVIĆ, 2016) and on the basis of the discriminant function analysis of the obtained mineral composition (GALOVIĆ & PEH, 2016), it can be concluded that the parent material of the studied recent soils has a similar mineral composition. The authors note that the modal composition in the areas covered by this work points to the Danube floodplain region (THAMÓ-BOZSÓ & KOVÁCS, 2007) and redeposited loess from Hungary (THAMÓ-BOZSÓ et al., 2014; ÚJVÁRI et al., 2008, 2014, 2016) as the main source of the material. As the Danube originates from the same region as the Sava and Drava rivers (the Alpine region), its mineral composition is similar.

However, as GALOVIĆ (2016) noted, the analysis of the modal composition of the loess horizons showed significant differences in the proportion of phyllosilicates (muscovite, biotite and chlorite), indicating the different area of origin and provenance of the source material. The source material for profile P-10 (rich in muscovite and other phyllosilicates) could be regional Tertiary sediments. These sediments are partly derived from the Paleozoic muscovite-rich rocks (e.g. garnet-muscovite gneisses, biotite gneisses and muscovite-biotite gneisses, mica schists, pegmatites and aplites) of the Slavonian Mountains (KOVAČIĆ et al., 2011; SLOVENEC et al., 2020; ŠEGVIĆ et al., 2022). Higher amounts of chemically stable muscovite indicate the possibility of multiple cycles of repeated resedimentation (FRECHEN et al., 2003; GALOVIĆ, 2014, 2016). These conclusions are also sup-

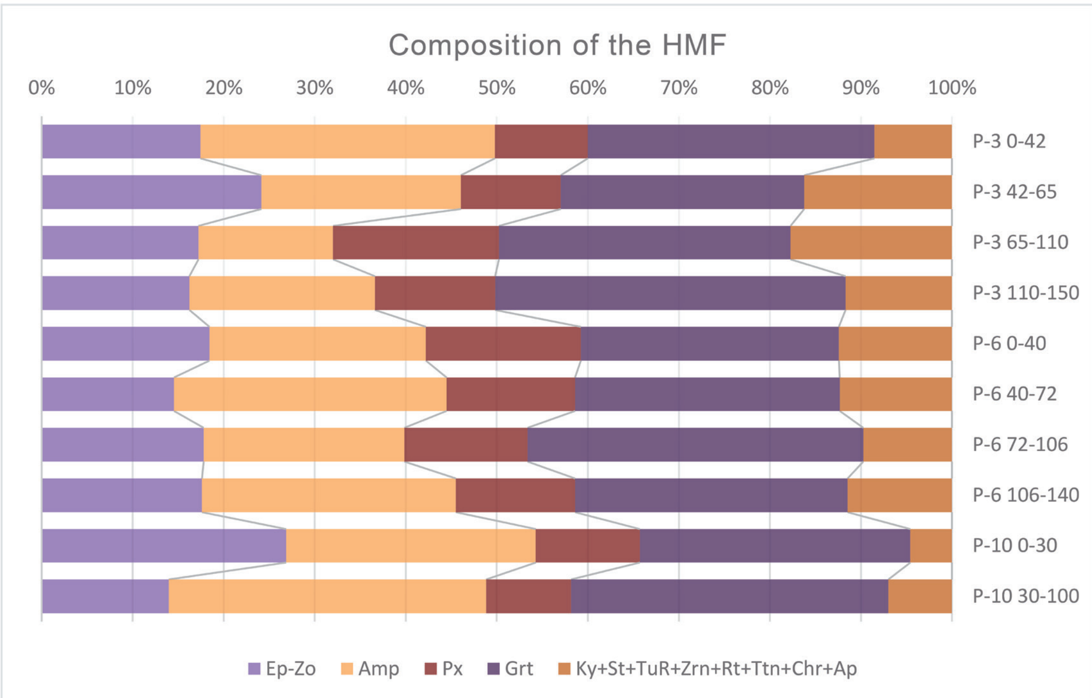


**Figure 12.** Modal composition of HMF without muscovite (values in %). Legend in Figure 8.

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ported by the frequent presence of regeneration rims on the weathered quartz grains in all investigated profiles. KLEPIKOV et al. (2022) described the mechanism of tangential growth from existing surfaces and concluded that regeneration growth is very rapid due, to the absence of the limiting nucleation stage of a new atomic layer (KEMPE, 2012). Thus, Alpine and local source areas are the sources of the aeolian sediments in the Pannonian Basin. However, the proportion and habitus of muscovite and other phyllosilicates indicate different local origins. Namely, soil profiles P-3 and P-6 contain well-rounded, smooth muscovite flakes, whereas the flakes from profile P-10 have fissured margins and contain a lot of needle-like inclusions. GALOVIĆ (2016) described in detail possible causes and sources for the enrichment of muscovite (and other flaky minerals). She claimed that the flat shape significantly favours aeolian transport. Namely, there is a possibility that the wind force that caused the accumulation of muscovite in the sediments was too weak to transport large quantities of uniformly sized and elongated grains. Modal compositions of loess in Croatia are presented in many publications (MUTIĆ, 1990; DURN, 2003; DURN et al., 2007; RUBINIĆ et al., 2015, 2018; WACHA et al., 2013). They indicate a homogeneous and uniform composition of the loess dominated by quartz and containing muscovite as a component of LMF. Since the muscovite-rich layers have a homogeneous composition along the P-10 profile, it can be concluded that there was no enrichment after the deposition of eolian sediments. The muscovite- rich material was homogenised before or during aeolian transport and then deposited in Zmajevac as homogeneous sediment before the onset of pedogenetic processes. Fractionation by wind energy in semi-arid mid-latitude regions during the Late Pleistocene could be influenced by both the aerodynamic properties of the particles (surface, shape) and density (ÚJVÁRI et al., 2016). Muscovite is resistant to chemical weathering and inert during pedogenesis. However, the unusual enrichment of muscovite led to a depletion of all other components of the transparent minerals of the HMF. In a "compositional dataset", changes in the content of one variable in a sample are accompanied by changes in the contents of other variables in the same sample (AITCHISON, 1986, 1997; GALOVIĆ, 2016; GALOVIĆ & PEH, 2016, in press).

Therefore, to enable a comparison of the composition of the transparent heavy minerals in the investigated profiles, an additional diagram was created that excludes the proportion of muscovite (Fig. 12). Figure 12 shows the enrichment of chemically (pedogenetically) not resistant amphibole in the uppermost horizon of profile P-3.

GALOVIĆ (2016) claims that abrasion dominates over chemical weathering because less weathering-resistant minerals (e.g. amphibole) are present in all studied horizons. However, in this research, the intense weathering of unstable mineral grains is confirmed by the chloritisation of amphibole and biotite. Although the presence of primary chlorite crystals can be proven by the occurrence of primary chlorite in lithic fragments originating from the mica schists, it was difficult to distinguish whether some particles were primary biotite or chloritised biotite. The decision was based on the degree of chloritisation of the biotite.

Detailed modal analyses of the LMF indicate that there are twice as many fresh quartz grains as weathered ones in soil profiles P-3 and P-6, while they are equally represented in profile P-10. Furthermore, weathered feldspar grains predominate in the uppermost horizons of profiles P-3 and P-6, especially in the uppermost horizon of profile P-3. There are twice as many fresh grains compared to weathered grains. In the P-10 profile, fresh and weathered grains are equally represented. The higher proportion of fresh grains in the surface horizons can also be confirmed by the modal composition of the HMF. The W.I. (ratio of chemically resistant and non-resistant transparent heavy minerals) shows an increasing trend with depth of the investigated profiles (Table 5; Fig. 9). A higher W.I. indicates significant or repeated weathering of the analyzed grains due to long exposure to a warm and humid geochemical environment and/or redeposition (FAIVRE et al., 2019). The analyzed soils are poorly developed compared to very well-developed Mediterranean soils, with a W.I. between 5.11 and 11.48% (FAIVRE et al., 2019).

The dominance of unstable minerals (epidote-zoisite group, amphibole, pyroxene and kyanite) in the surface horizons and the tendency for their proportion to decrease with depth indicate a continuous eolian enrichment of the soil with fresh, unweathered material. The new eolian contribution was introduced into the soil profiles naturally (profile P-10) and/or by human action (profiles P-3 and P-6). The evidence for synpedological aeolian sedimentation is the higher content of fresh quartz and feldspar grains, sporadic spherical grains of devitrified volcanic glass and weathered dolomite grains in the Ap horizons of profiles P-3 and P-6. The eolian contribution in recent soil profiles is described in GALOVIĆ & PEH (2014). FAIVRE et al. (2019) assumed that the Milna drainage basin have a polygenetic origin, similar to terra rossa along the eastern Adriatic coast (DURN et al., 2007; ROMIĆ et al., 2014). Furthermore, a recent investigation of loess sections in the south-eastern and central Carpathian Basin (MARKOVIĆ et al., 2023) indicates that favourable local vegetation conditions for dust uptake existed throughout the Late Pleistocene. These environmental conditions promote higher availability of source material for further aeolian activities. The present records provide new insights into dust accumulation regimes over the eastern side of the Bačka loess plateau. They represent an important step towards the establishment of a chain line from the thin loess-like sediments of the Banat foothills in the east to the thicker and apparently more complete loess sections of the south-eastern and central Carpathian Basin.

In addition to the modal analysis, the composition of the clay fraction also suggests that the parent material of investigated soil profiles is underlying loess-paleosol sequences, the mineralogical composition of which is also consistent with some previous works (GALOVIĆ, 2016; GRIZELJ et al., 2016; URUMOVIĆ et al., 2017). According to them, the mineralogical composition of the underlying loess-paleosol sequence consists of quartz, micaceous minerals (illite and muscovite), chlorites and expandable clay minerals, while to a lesser extent it contains kaolinite, carbonates and feldspars. A very similar composition was obtained by analysis of the soil samples from profiles P-3 and P-6. Quartz, micaceous minerals and calcite predominate in bulk composition, which is also the case in the parent material. In the fraction < 2 μm of soil profiles P-3 and P-6, the dominant clay minerals are illite and chlorite, whose content is constant in all horizons. They most likely originate from the parent material, especially the chlorite grains present in the silt fraction. Modal analysis revealed that a small proportion of chlorite was formed by the chloritisation of biotite and amphiboles. Similar to chlorite, vermiculite can also be formed by the decomposition of biotite or by the decomposition of chlorite during pedogenesis (VELDE & MEUNIER, 2008). This is indicated by the presence of chlorite mixed layer clay minerals in the C horizons of profile P-6. Slightly lower amounts of vermiculite in the upper part of both profiles could indicate the displacement of the clay fraction by water infiltration and wind erosion (ALTAY, 1997) or the transformation of a vermiculite clay mineral to kaolinite.

In well developed soils, the end product of weathering of soil minerals is kaolinite or smectite together with iron oxides and hydroxides. In analyzed soils, the presence of well crystallized kaolinite is clearly detectable and most likely inherited. There is no clear evidence of the presence of poorly crystallized kaolinite in the samples. Although modal analysis revealed the presence of feldspar grains with weathered rime that could consist of poorly crystallized kaolinite the 7 Å kaolinite peak is not present in the x-ray diffraction analysis. The reason for this may be the small amount of poorly crystallized kaolinite, but also the presence of chlorite and vermiculite, whose reflection at the same diffraction position masks the reflection of kaolinite. The presence of kaolinite, even in very small quantities, as well as goethite, may indicate an influence of chemical weathering on the degradation of chernozem profiles. In this case, it cannot be claimed with certainty that the feldspars have been weathered recently, primarily due to the basic environment with a pH of > 8 (Table 4).

## 5. CONCLUSIONS

The research results confirm the existence of chernozem in Croatia, both according to the Soil Classification of Croatia and the World Reference Base for Soil Resources. Considering the area occupied by the chernozem soil type according to the Croatian soil classification, which is about 50,000 ha, it cannot be claimed on the basis of these studies that the entire chernozem area belongs to the reference group of chernozem soils according to the Word Reference Base for Soil Resources. Based on soil characteristics, changes indicating soil degradation have been identified, in particular the reduction of organic matter and the displacement of carbonates from the surface to deeper zones. The presence of a chernozem degradation process due to the impact of recent weathering processes can only be indicated by the presence of goethite in the fraction < 2 µm. Kaolinite and weathered feldspar grains are not products of recent weathering, primarily due to the alkaline environment, but are most likely inherited.

Although the parent material of the studied recent soils has a similar mineral composition, differences in the proportion of phyllosilicates (muscovite, biotite and chlorite) and their habitus indicate the different area of origin and provenance of the source material. Furthermore, the dominance of unstable minerals (epidote-zoisite group, amphibole, pyroxene and kyanite) in the surface horizons and the tendency for their proportion to decrease with depth indicate a continuous eolian enrichment of the soil with fresh, unweathered material. The synpedological eolian sedimentation is confirmed by the higher content of fresh quartz and feldspar grains, sporadic spherical grains of devitrified volcanic glass and weathered dolomite grains in the Ap horizons of profiles P-3 and P-6.

Following the principle of actualism, it is therefore important to be aware, during investigations of paleosols, even buried ones, that synpaleopedological aeolian sedimentation could occur and lower a certain degree of pedogenesis.

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