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The Late Pleistocene Belotinac section (southern Serbia) at the southern limit of the European loess belt: Environmental and climate reconstruction using grain size and stable C and N isotopes



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ABSTRACT

The Belotinac loess section is one of the southernmost loess-paleosol environmental archives for the Late Ouaternary in Serbia. The climate at this site is intermediate between the continental and the Mediterranean realms, which makes this loess archive potentially highly sensitive to past climatic changes. This paper presents new insights into the paleoenvironmental history during the last glacial period in southern Serbia using grain size and isotope proxy data. The grain size parameters from the Belotinac section reveal variations in the paleowind dynamics and weathering intensity, and suggest the nearby valley of the Južna Morava River as an important source of aeolian sediments in this area. Based on a multiproxy dataset, alternating phases of weak interstadials and phases of enhanced loess deposition at this site were identified. Nitrogen isotope data suggest that during Marine Isotope Stage 3, ecosystems of high biomass productivity and rather open N-cycles prevailed. During Marine Isotope Stage 2, productivity was reduced and the N cycle was more strongly closed, probably due to a shorter growing season and more pronounced temperature decline. Carbon isotope data indicate a possible contribution of C4 plants to the Holocene vegetation, but not to the glacial and interstadial ecosystems of the Late Pleistocene. Changes in atmospheric CO₂ level are not reflected in the carbon isotope record. These findings are discussed in the light of paleoclimate proxy datasets from the Morava River valley and Carpathian Basin, as well as through comparison of carbon isotope records from loess sections in SE - Central Europe and in the Rhine Valley. Two different loess provinces exist in terms of glacial-interglacial humidity changes: a province of "glacial drying" and a province of "glacial humidification". The first includes loess sites under more oceanic influence, where loess δ^{13} C records indicate humid interglacials and interstadials and relatively drier glacial periods. The second includes the loess sites of the Carpathian Basin and especially the southern Serbian loess area of Belotinac, where loess δ^{13} C records indicate more intensive aridity during interglacials, but a reduced soil moisture deficit and a more humid climate due to lower evapotranspiration in interstadials, and even more in glacial periods.

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1. Introduction

* Corresponding author. *E-mail address:* obrehtigor@gmail.com (I. Obreht). Terrestrial wind-blown sediments provide sensitive records of past climatic and environmental change (Stevens et al., 2008; Stevens and Lu, 2009). Among these, loess-paleosol sequences are

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widespread on the continents and can preserve detailed evidence of different natural processes and their dynamics (Smalley et al., 2011).

In contrast to the well studied exposures in the northern Serbian province of Vojvodina (Markovič et al., 2005, 2006, 2007, 2008, 2009, 2011, 2012; Buggle et al., 2009, 2011; Újvári et al., 2010; Stevens et al., 2011; Liu et al., 2013), loess sections in other Serbian regions have not been investigated intensively (Kostić and Protić, 2000; Mitrović and Jovanovič, 2000; Mitrović, 2004). To close this gap, we started multidisciplinary investigations of the Belotinac site in southern Serbia (Basarin et al., 2011).

Present climate conditions in the Vojvodina region and the area around Belotinac (climate station of Niš city) are quite similar (Ducić and Radovanović, 2005) and are suitable for the development of a forest-steppe mosaic vegetation cover. However, southern Serbia experiences a stronger Mediterranean climate influence, causing higher summer and winter air temperatures than at Vojvodinian loess sites. Loess sites in southern Serbia such as Belotinac characterize the transition zone between the Mediterranean and continental realms, and potentially are extraordinarily sensitive to past and present climate change.

In this study, we applied grain size and geochemical investigations to reconstruct climate and environmental dynamics. Grain size composition is one of the most frequently used paleoenvironmental proxies of loess deposits to infer changes in aeolian dynamics, loess source and pedogenesis (Vandenberghe et al., 1997; Ding et al., 2002; Prins et al., 2007; Bokhorst et al., 2011). This multidisciplinary study also interprets carbon and nitrogen stable isotopes (c.f. Zech et al., 2007; Schatz et al., 2011), presenting a combination of the classical paleopedological—sedimentogical proxies and stable isotope-based palaeoecological proxies. The results are closely linked to a previous study (Basarin et al., 2011), and aim to reconstruct environmental changes during the last 35,000 years in southern Serbia.

2. Material and methods

2.1. Belotinac section – stratigraphy and chronology

The Belotinac section (43°15.889'N; 21°52.163'E, 228 m a.s.l.) is exposed in an abandoned quarry (10 km southwest of Niš), on a river terrace at the eastern side of Južna Morava River valley (Fig. 1). At present, the mean annual temperature and precipitation from the nearby climate station at Niš are about 11.4 °C and 589.6 mm (with the main precipitation maximum during June and May, and a second maximum during November and December). Mean July and January temperatures are 21.2 °C and 0.2 °C, respectively.

A detailed description of the Belotinac section and stratigraphic interpretation was presented by Basarin et al. (2011). Fig. 2 represents the main stratigraphic units of the exposure. Using lithologic and pedologic criteria as well as preliminary chronological data, Basarin et al. (2011) established the first chronostratigraphic model for the Belotinac loess-paleosol sequence.

The profile has a total thickness of 8 m. In the lowermost part, a sandy layer with a thickness of approximately 2 m is found. The sandy sediments at the base of the section are overlain by a chernozem-like paleosol L1S1, which is 125 cm thick. On top of unit L1S1, there is a thin loess layer L1L1L3, approximately 25 cm thick. This loess layer is overlain by a darker thin layer, which can be considered as a weakly developed chernozem-like paleosol, L1L1S2. Above L1L1S2, a loess layer L1L1L2 was deposited, 50 cm thick, with similar characteristics as the loess stratum L1L1L3. Some secondary calcite deposits (pseudomycelia) in the pores were observed in L1L1L2. Paleosol unit L1L1S1, 120 cm thick, is superimposed over loess layer L1L1L2. The uppermost loess layer L1L1L1 is 130 cm thick and consists of typical porous loess. A 100 cm thick modern

cambisol, S0, represent the top of the sequence. It comprises a strongly developed (7.5 YR 5/6) B horizon, followed by an AB horizon (7.5 YR 4/4) and an Ah horizon (7.5 YR 4/4, 7.5 YR 5/4) on top.

Previously published Infrared Stimulation Luminescence (IRSL) ages of the Belotinac site range from 22 ± 2 ka to 35 ± 4 ka (Fig. 2). Although the dataset is limited, the dates allowed Basarin et al. (2011) to establish a broad chronostratigraphic framework. The IRSL chronology allocates the loess-paleosol sequence at Belotinac to MIS2 and 3 (Basarin et al., 2011).

The section was sampled at 10 cm intervals for grain size and geochemical analysis. Additionally, samples were collected for measurements of magnetic susceptibility, frequency dependent magnetic susceptibility, color measurements, and luminescence dating (Basarin et al., 2011).

For grain size measurements, sub-samples of 5 g sediment were decalcified (2 M HCl), dispersed with sodium-hexa-metaphosphate $((NaPO_3)_6)$ and stored >12 h in an overhead shaker. The fractions $>63 \mu m$, $>200 \mu m$, and $>630 \mu m$ were determined through sieve analyses. The $<63 \,\mu m$ fraction was measured in the laboratory of the Chair of Soil Physics, University of Bayreuth, on a Malvern Mastersizer S (Malvern Instruments, Worcestershire, UK) with a measurement range of 0.02 µm-2 mm. Measurements were conducted in duplicate analyses to ensure the reproducibility of the results. The U ratio, the ratio of the 16-44 and 5.5-16 µm fractions, is a grain sizebased paleoenvironmental proxy. This ratio is well established in aeolian sediment research, as it primarily reflects changes in the grain size distribution due to varying sedimentological conditions, giving information on wind strength. It disregards secondary minerals formed during pedogenesis in the clav fraction, and sand-sized particles probably deposited by saltation (Vandenberghe et al., 1997).

As proxy for the clay content, we used the $<5 \mu$ m laser grain size fraction (Buggle et al., 2013). This is in agreement with other published clay fraction equivalent values for Serbian sections: $<4.6 \mu$ m by Antoine et al. (2009) and $<5.5 \mu$ m by Bokhorst et al. (2009), determined on various types of laser particle size analyzers. The $<5 \mu$ m size fraction is predominantly pedogeneticallyformed particles and hence should reflect pedogenic intensity. Nevertheless, there might be a contribution of detrital clay particles originating from the dust source region. This has been observed in Chinese loess (Yang and Ding, 2004), but cannot be quantified here.

For the determination of the total organic carbon content (TOC), total nitrogen content (TN) and the stable isotope signature of organic carbon and nitrogen, samples were pretreated according to the procedure proposed by Gauthier and Hatté (2008) to remove inorganic carbon. In brief, aliquots of about 200 mg per sample were leached with 0.6 N HCl in preashed glass vials for 5 days at 40 °C, daily refreshing the acid. The acid supernatant was carefully sucked off using Pasteur pipettes, and samples were carefully rinsed several times with ultrapure water to remove remaining chloride ions. After settling, the aqueous supernatants were again sucked off and the samples were dried at 40 °C. Tests with carbonate-free reference material spiked with 20% dolomite confirmed that this procedure is suitable for removing inorganic carbon, and minimizes loss of organic compounds due to hydrolytic reactions. TOC, TN, δ^{13} C and δ^{15} N were determined using dry combustion of ~40 mg subsamples with a Carlo Erba NC 2500 elemental analyzer coupled to a Delta Plus continuous flow isotope ratio mass spectrometer (IR-MS) via a Conflov II interface (Thermo Finnigan MAT, Bremen, Germany).

3. Results

3.1. Granulometric composition and proxies

This study presents the results of grain size variations recorded in the Belotinac section. The typical grain size distribution pattern



Fig. 1. Map of SE Europe and the location of Belotinac loess-paleosol sequence. Legend: 1. Contour lines; 2. Alluvial sediments; 3. Settlement; 4. Road; 5. Elevation above sea level.

of the individual stratigraphic units (S0, L1L1L1, L1L1S1, L1S1 and basal sandy horizon) is shown in Fig. 2. High resolution downprofile records of the individual grain size fractions and the U ratio is given in Fig. 3.

The silt fractions make a significant contribution to the grain size inventory from the main part of the section from L1S1 to the top, reflecting the input of aeolian dust to the sedimentary sequence. Significant contributions of coarse silt and fine and medium sand fraction (between 51 and 66%) are observed in all analyzed samples. The content of fine sand generally increases with depth, from less than ~35% in the upper part of the profile to more than 50% in the basal layer below L1S1. The coarse and medium sand fraction (>200 μ m) shows a strong variability throughout the profile, with a slight increase with depth. In the basal layer, particles with mainly pedogenetic origin (<5 μ m) and dust particles (5–60 μ m) represent only minor fractions, and sand dominates the suite of grain size fractions. These grain size patterns reflect the

lithological structure and genesis of the archive (Basarin et al., 2011), as the loess paleosol sequence from L1S1 to the top is developed on a stratum of fluvial sand originating from the nearby Južna Morava River.

The clay content ($<5 \ \mu$ m) fraction is also consistent with the lithostratigraphy (Basarin et al., 2011), with the highest values in the modern soil (up to 25%). However, at Belotinac there is no pronounced clay enrichment in the interstadial paleosols as compared to the underlying loess units. The $<5 \ \mu$ m record shows high variability within individual pedocomplexes, indicating multiple periods of weathering interrupted by periods of high dust input. A high variability related to the sedimentary system is also indicated by the U-ratio. High frequency oscillations in this parameter inversely correlate with small scale changes in the clay content (excursions to lower U ratios correspond to subtle peaks in the clay content), suggesting a close connection between sedimentation and weathering/clay formation at Belotinac. At profile



Fig. 2. Lithostratigraphy of the Belotinac loess-paleosol sequence including stratigraphic position of luminescence dating results (Basarin et al., 2011) and typical grain size distribution patterns of the individual stratigraphic units.

scale, however, the U-ratio shows a distinct shift to lower values from 2.5 to 6 m profile depth, consistent with the lithostratigrapic units of weak soil formation within the L1 as determined by soil structure and colour (Basarin et al., 2011). This suggests coupling of dust sedimentation and pedogenesis, with pedogenesis being in phase with a low capacity of the aeolian system in terms of deflation and transport. 3.2. C and N elemental concentrations and stable isotopic compositions

3.2.1. Total organic carbon (TOC) and total nitrogen (TN)

The results of TOC analysis show enrichment of organic carbon in the modern soil and paleosols, with low values observed in loess units (Fig. 4). The lowest TOC values are recorded in the basal sandy unit with a gradual decrease from the top to the base. TOC variations in paleosol L1S1 are characterized by two prominent peaks with values of around 3%, similar to the modern soil SO. The TOC values are reported on a carbonate-free basis, and therefore are generally higher than values normalized on bulk material (including carbonates), previously determined on selected samples by Basarin et al. (2011). High TOC values in the modern as well as fossil soil indicate a dry and/or a seasonally cold climate, hampering the degradation of organic material. Pulsed phases of high sedimentation rates, as potentially indicated by the high frequency fluctuations in clay content and the U ratio, may favour organic material preservation by burial. TOC values of other loess and paleosol units are in the range of 0.7–2.0%. The weakly developed paleosols such as L1L1S1 and L1L1S2 can be clearly distinguished from the loess using the elevated TOC values. Hence, increased biomass incorporation in the loess is coupled with periods of enhanced soil-forming intensity, potentially related to the intensity of biomass production. However, in loess-palaeosol sequences, this might also relate to changes in the balance between aeolian deposition and biomass production and preservation. The two pronounced peaks in TOC found in unit L1S1 indicate a multi-phase evolution of the interstadial pedocomplex, in terms of changing climatic conditions (shifts in biomass production) and/or varying synpedogenic sedimentation rates (varying dilution of organic by inorganic flux).

Total nitrogen is highly correlated with the TOC. This indicates a common biotic source (biomass input from ancient ecosystems) and highlights the potential of combined analyses of stable nitrogen and carbon isotopes to characterize past ecological conditions.

3.2.2. Stable carbon isotopes and nitrogen isotopes

The δ^{13} C values recorded at the Belotinac profile range approximately from -21 to -25% (Fig. 4), with the lowest values in the sandy layer at the base of L1S1. However, a slight trend of δ^{13} C enrichment from bottom to top in the profile is evident, with values



Fig. 3. Magnetic susceptibility, colour reflectance (a*chromaticity) (Basarin et al., 2011) and grain size proxies related to pedostratigraphy. Ages shown in ka next to the sequence represent the results of luminescence dating. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 4. Magnetic susceptibility, colour reflectance (a*chromaticity) (Basarin et al., 2011) and geochemical proxies related to pedostratigraphy. Ages shown in ka next to the sequence represent the results of luminescence dating. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of ~ -24 to -23.5‰ in the oldest paleosol L1S1, and up to -23‰ in the loess of the last glacial maximum. A distinct shift towards more positive values (-21‰) appears at the base of the modern soil.

Fig. 4 also shows the δ^{15} N record. The values cover a range from 2 to 6‰. Pedogenetic layers are characterised by more positive δ^{15} N values compared to loess layers, with similar values in the modern soil and the fossil soils. Some distinct peaks of relatively high δ^{15} N values (up to 8‰) occur, not only in soil layers but also in intercalated loess layers. This feature is not regarded as a paleoenvironmental signal, but as a methodological artefact due to the sample preparation (acid treatment procedure, e.g. Ryba and Burgess, 2002; Kennedy et al., 2005).

4. Discussion

4.1. Grain size and mineralogical perspective on the Late Pleistocene environment of Belotinac

The grain size record of the Belotinac section indicates only a slight increase in pedogenic clay formation in paleosol L1S1, which is masked by many high frequency oscillations. In the other weak interstadial paleosol units L1L1S1 and L1L1S2, the pedogenic <5 μ m fraction is not higher. As the intensity of pedogenic clay formation does not reflect the lithostratigraphy, the <5 μ m fraction cannot be applied as a climate proxy at Belotinac. However, paleoenvironmental implications may be drawn from the absence of pedogenic clay formation in the fossil interstadial soils.

High resolution grain size records from loess paleosol sequences in Vojvodina such as the Titel old brickyard (Bokhorst et al., 2009) and Surduk sites (Antoine et al., 2009) generally show a distinct increase of the pedogenic clay fraction in interstadial soils of the Late Pleistocene, on the order of $\sim 5-15\%$. In this study, the grain size record is also clearly correlated with the magnetic susceptibility as an independent proxy for pedogenesis. Fig. 3 compares the lithology and grain size parameters of Belotinac with the records of magnetic susceptibility, frequency dependent magnetic susceptibility and sediment color presented by Basarin et al. (2011) for the Belotinac sequence. Although the magnetic susceptibility gives an integrated signal from all ferrimagnetic particles, the frequency dependent susceptibility is sensitive only for small ferrimagnetic particles close to the threshold of superparamagnetic to single domain behavior (Banerjee, 1994; Liu et al., 2007). This broadly represents the fraction of ferrimagnetic minerals formed during pedogenesis. Although the frequency dependent magnetic susceptibility shows a general increase in the units L1S1-L1L1S1 as compared to the basal layer and the LGM loess (unit L1L1L1), the intensity is quite low, compared with the modern soil. Moreover, the frequency dependent susceptibility does not reflect lithological changes between the interstadial soil units and the intercalated loess units (L1L1L3 and L1L1L2). This suggests that climatic conditions required for the pedogenic formation of ferrimagnetics did not vary much between these units. The strong enhancement in the bulk magnetic susceptibilities characterizing the paleosol units L1S1 and L1L1S1, in contrast, are not related to pedogenesis but likely to varying amounts of highly susceptible lithogenic ferrimagnetic minerals in the dust (see Basarin et al., 2011 for an extensive discussion of the rock magnetic parameters). Hence, the magnetic susceptibility record is a proxy coupled to the sedimentary system with varying dust sources. Due to the relatively high contribution of sand and coarse silt, the potential major source for aeolian sediments at Belotinac is sediment from the nearby Južna Morava River. Magnetic susceptibility is about one order of magnitude higher than in loess deposits along the Danube, at the Eastern and Northern shores of the Black Sea and in Central Asia and China (e.g. Buggle et al., 2009; Markovič et al., 2009). Hence, rock magnetic properties of the Belotinac loess-paleosol sequence are primarily controlled by the geology in the Južna Morava catchment, characterized by metamorphic and igneous rocks that provided minerals with high magnetic susceptibility (Basarin et al., 2011). The importance of fluvial sediments as dust sources has been previously confirmed by Buggle et al. (2008) and Újvári et al. (2008) for loess sites in the Carpathian Basin and in Romania. At Belotinac, there is likely a second dust source of unknown origin, with a different load of ferrimagnetic minerals. This source appears to be of enhanced significance in periods of soil formation (L1L1S1, L1S1). This suggests that formation of Late Pleistocene interstadial soils at Belotinac is intimately controlled by the intensity of aeolian deposition. Climate conditions likely were suitable to allow weak pedogenesis throughout the units L1S1 up to the base of LGM loess L1L1L1, indicated by the frequency dependent susceptibility. Formation of well-developed interstadial soil layers (L1S1, L1L1S2, L1L1S1), however, is restricted to periods of reduced dust deposition. Consequently, units L1L1L3, L1L1L2 and L1L1 represent periods of high aeolian activity, in which imprints of the organic flux from *in situ* biomass production and the iron pigment flux from brunification has been overridden by enhanced aeolian deposition.

4.2. Late Pleistocene environment of Belotinac from an isotope perspective

4.2.1. Carbon isotopes and degradational effects

 δ^{13} C values in (fossil) soils and terrestrial sediment can reflect paleoenvironmental conditions, but they also might be affected by changes in the isotopic composition due to degradation. Therefore, the significance of a potential degradational bias of the isotope record has to be assessed. The δ^{13} C values at the Late Pleistocene loess-paleosol successions of Belotinac are within the range of \sim -23 and \sim -24%. These values are slightly more positive than would be expected for typical C₃ vegetation (~ -27 to -25%). Such isotope enrichment could be an effect of organic matter degradation (e.g. Wynn et al., 2006). Degradation intensity of organic matter is generally assumed to differ between loess units (such as L1L1L1, L1L1L2 and L1L1L3) formed in glacial environments with high sedimentation rates, and paleosols formed in interstadial environments (such as L1L1S1, L1L1S2 and L1S1). The absence of any systematic enrichment in the paleosols as compared to the loess units suggests that degradational effects did not significantly alter the carbon isotope values in the Late Pleistocene part of the Belotinac profile. Hatté et al. (1998, 1999) stated that the δ^{13} C signal is not effectively altered by degradation in loess units and weak interstadial soils in loess sites in the Rhine valley. In contrast, in more strongly developed interglacial soils such as the Holocene soil of Belotinac, degradational effects can contribute to an enriched δ^{13} C signal. In the lower parts of the Holocene soil, carbon isotope values of -21% might indicate a contribution of C₄ plants. This value, however, represents an enrichment of only 1% compared to the modern surface of this soil. Regardless of any vegetation change, isotope enrichment due to organic matter degradation in soils of temperate and semi-arid environments are documented to be typically between 1 and 3% (Krull et al., 2006 and references therein). Some studies report isotope enrichments of up to 4 and 5% (Krull et al., 2006; Alewell et al., 2011 and references therein) in extratropical soils, where $C_3 - C_4$ vegetation changes can be excluded. Hence, it is not possible to unravel a possible contribution of C₄ plants from a degradation effect when interpreting the positive carbon isotope excursion in the Holocene soil. A "degradation effect" scenario might fully explain the observed isotope shifts. However, a C₄ contribution cannot be disproved by these isotope data.

4.2.2. Carbon isotopes and environmental factors

In addition to degradation effects, factors potentially controlling isotope ratios in soils and sediments are changes in the photosynthetic pathway (shifts from C₃ to C₄ vegetation), physiological water stress, and changes in the atmospheric CO₂ concentration (O'Leary, 1988; Hatté et al., 1999). Major changes from a C₃ to a C₄ plant community generally induce large amplitude changes, on the order of more than 9‰ enrichment of δ^{13} C in the organic material (Boutton et al., 1998). Such changes were not observed at Belotinac. For the Late Pleistocene sediment soil successions, the measured δ^{13} C signature is within the range of C₃ vegetation.

The absence of evidence for C_4 vegetation in this area during glacial times has paleoclimatic implications. Species using the C_4 photosynthetic pathway have higher water use efficiency and a higher temperature optimum for net primary production (Ehleringer, 1978; O'Leary, 1981; Ehleringer et al., 1997; Collatz et al., 1998; Hatté et al., 1999, 2001; Kohn, 2010). Higher

atmospheric CO₂ level, lower temperature, and higher rainfall favor C₃ plants over C₄ plants (Sage et al., 1999). Temperature effects generally dominate over changes in precipitation (Hall et al., 2012). Consequently, transitions from C₃ to C₄ vegetation cannot be universally interpreted as evidence of decreased precipitation.

The crossover temperature for the dominance of C_4 species is related to the atmospheric CO_2 level, and is today about 22 °C (Ehleringer et al., 1997; Collatz et al., 1998), as related to the mean temperature of growing season months. In Mediterranean and Mediterranean-like (Yang and Ding, 2006) climates with the main growing season in winter and spring, C_3 vegetation dominates. In the Carpathian Basin steppe, the main growing season under modern climate conditions is May and June, coeval with the predominant period of precipitation. For these months, mean temperatures from the climate station Niš, near Belotinac, were between 16.6 °C and 19.5 °C (http://www.hidmet.gov.rs), well below the crossover temperature. Collins and Jones (1985) reported a C_4 contribution of less than 2% to the modern flora of this region.

For pre-industrial CO₂ levels of 270 ppm, the crossover temperature for C₄ vegetation was 18 °C (c.f. Collatz et al., 1998). Accordingly, significant abundance of C₄ vegetation in the flora of the Holocene could be expected in this region. The lower part of the Holocene soil shows carbon isotope values to $\sim -21\%\sigma$ These values typically reflect a C₃ dominant – C₄ subordinate vegetation. The expansion of C₄ plants observed in the lower part of the Holocene soil might represent an imprint from ecosystems adapted to a pre-industrial CO₂ world, possibly combined with an imprint from the Holocene climate optimum.

At atmospheric CO₂ levels of ~180 ppm, as during the last glacial maximum, or ~220–240 ppm, as during interstadials of the last ~40 ka (Jouzel et al., 1993), crossover temperatures would be much lower, about 10 °C and 12–13 °C, respectively. During glacial conditions, summer months (June–August) most probably represented the major part of the growing season. Full-glacial mollusc assemblages in the central and southern part of the Carpathian Basin indicate that the July mean temperature was more than 12 °C, and on south facing slopes more than 17 °C (Sümegi and Krolopp, 2002; Markovič et al., 2007). Hence, growing season temperatures at Belotinac should have been well above the crossover temperature during the Last Pleniglacial.

Changes in the atmospheric CO₂ level also influence the carbon isotope signature of C₃ species. Feng and Epstein (1995) determined the response of δ^{13} C in tree rings to changes in the atmospheric CO₂ concentration to be -0.02%/1 ppm CO₂. Taking this value as approximation for the sensitivity of fossil organic matter δ^{13} C values to record past changes in atmospheric CO₂, would give an estimated isotopic difference between pre-industrial Holocene and LGM values of about 2% (c.f. Hatté et al., 1998). At Belotinac, a similar feature was not found in the youngest loess unit (L1L1L1), nor at the transition to the Holocene soil. The ~20–30 ka units also do not reveal any substantial variation in the carbon isotope record.

Despite the apparent suitability of environmental conditions, the δ^{13} C record does not show evidence for C₄ vegetation in any of the loess units. This finding is consistent with the absence of evidence for C₃ to C₄ vegetation changes in other published δ^{13} C loess records from sites of the Carpathian Basin (Schatz et al., 2011; Zech et al., 2013).

A possible explanation for the discrepancy is the presence of trees during glacial conditions. Charcoal (Willis et al., 2000; Willis and Van Andel, 2004) and n-alkane biomarker studies (Zech et al., 2009) indicated that the relative abundance of trees in glacial periods was higher than during interstadials or interglacials, the result of reduced water stress and higher soil moisture. Steppe (C_4 dominated) vegetation was favoured under interglacial conditions with lower soil moisture. The increase in tree (C_3) abundance

could have counterbalanced the increasing abundance of C₄ vegetation, influencing the isotopic signature in the last glacial loess.

4.2.3. Tracking last glacial carbon isotope records in Central European loess

The published carbon isotope records from European loess include the Crvenka (Zech et al., 2013) records in Serbia, the Tokaj record in N-Hungary (Schatz et al., 2011), and the Nussloch and Achenheim records in W-Germany and E-France respectively (Hatté et al., 1998). Fig. 5 shows a direct comparison between δ^{13} C records obtained from the Achenheim, Nussloch, Tokaj, Crvenka, and Belotinac sections, plotted on a depth scale. Compared records cover different time frames from the last 35 ka to the entire Late Pleistocene. The most significant differences among the analyzed records are observed during the last glacial maximum period and the Holocene. δ^{13} C values range from approximately -27.5% at Achenheim to almost -21% at Belotinac. In the older parts of the records, δ^{13} C values show reduced variations, ranging between -25% and -23%, with the exception of several prominent peaks observed in the Achenheim and Tokaj profiles.

The carbon isotope records from Crvenka (Fig. 5; Zech et al., 2013) and the Tokaj record in part are very similar to that of Belotinac, showing almost constant values around -23 to -24.5% in stadial, interstadial, and glacial units. The Tokaj site is situated at the foothills of the northern Carpathian fringe, and today represents a more humid climate province of the Carpathian Basin. At this site, a marked excursion to -27% in the upper interstadial paleosol was found by Schatz et al. (2011). At Nussloch, the highest δ^{13} C values in glacial loess are in the range from -23.5 to -24% and at Achenheim from -22.5 to -23.5% In the Upper Pleniglacial

loess of Nussloch, small scale negative excursions of high frequency to values of about -24.5% to -25% appear to be correlated with weak periods of soil forming conditions during interstadials. A distinct shift of similar magnitude characterizes the Middle Pleniglacial soil complex at Nussloch (Fig. 5). These small scale excursions in the carbon isotope record have been interpreted as paleoprecipitation or soil moisture supply signals by Hatté et al. (1998, 1999, 2001) and Hatté and Guiot (2005), who correlated these features to D/O cycles. Hatté et al. (1998, 1999) found a decrease of δ^{13} C in the late glacial loess of Nussloch, which they relate to the rise in atmospheric CO₂. At Achenheim, the pattern of the $\delta^{13}C$ record less clearly reflects the stratigraphic context. This possibly might be a result of postpedogenic or postsedimentary rooting contamination. The potential significance of rooting contamination in organic proxy-based records has been previously highlighted by Gocke et al. (2010).

Notwithstanding the disturbed record of Achenheim, there appears to be a spatially systematic pattern in δ^{13} C records of the last glacial in Europe. The carbon isotope records from the loess sites in the Rhenisch loess area of Eastern France and Western Germany show a distinct increase in humidity during interstadials and interglacials as compared to glacials. During warm stages, climates in this area are more strongly influenced by oceanic air masses from the Atlantic Ocean. This feature is designated as the " δ^{13} C pattern of humid warm stages". In contrast, Crvenka in the southeastern Carpathian Basin and Belotinac in the central part of Balkan region show no systematic pattern indicating increasing humidity in interstadials. A decrease in δ^{13} C during interstadial periods should be expected in this area due to the plant physiological response to lower atmospheric CO₂ levels in the glacial phases. A dry



Fig. 5. Direct comparison between δ^{13} C records obtained from loess sections at Nussloch in Germany and Achenheim in France (Hatté et al., 1998), Tokaj in Hungary (Schatz et al., 2011; Zech et al., 2013), Crvenka (Zech et al., 2013) and Belotinac in Serbia are plotted on a depth scale.

interglacial/interstadial climate (compared to glacial conditions) is postulated for this area, and $\delta^{13}C$ records reveal a pattern of arid warm stages.

The Tokai site shows a clear increase of frequency dependent magnetic susceptibility in all interstadial pedocomplexes, reflecting higher temperature and precipitation values during the interglacials. In one paleosol at Tokai, the δ^{13} C record indicates an increase in humidity as a short negative excursion. Hence, the northern part of the Carpathian Basin and the foothills of the central Balkan mountain ranges region appears to be a transitional area between the arid warm stage region (province of glacial humidification) and the humid warm stage region (province of glacial drying). There is a need for more δ^{13} C records to validate the observed regional pattern. Nevertheless, besides these first indications from the isotope perspective, the validity of this pattern is underlined by paleoenvironmental evidence from charcoal and biomarker analyses in the Carpathian basin (Willis et al., 2000; Willis and Van Andel, 2004; Zech et al., 2009). These paleovegetation markers indicate expansion of trees due to reduced net evapotranspiration in the glacial periods.

4.3. Nitrogen isotopes and the influence of biomass productivity

The $\delta^{15}N$ record shows generally more positive values from \sim 2.2 m to 6 m depth in the Belotinac section. This part of the section covers interstadial paleosol units (L1S1, L1L1S2, L1L1S1) as well as intercalated loess lavers (L1L1L1, L1L1L2, L1L1L3). The intensity of nitrogen isotope enrichment in natural soils is predominantly an imprint of changes in the N-cycle, more specifically the openness and closeness of the N-cycle. Open N cycles are typical for ecosystems that are not N-limited due to high input rates of N and high mineralisation rates, and/or due to low uptake rates by plants. In open N-cycle environments, isotopically depleted inorganic nitrogen is preferentially released by nitrification or denitrification reactions and subsequently leached or degassed from the pedosphere (Krull and Skjemstad, 2003; Zech et al., 2011; Zech et al., 2013). Hence, isotopic enrichment of the remaining soil N can be taken as an indicator for open N-cycles, whereas there is only a small loss of isotopically depleted inorganic nitrogen in rather closed N-cycles (small input rates of N or high uptake rates). Similar isotopic enrichment has been observed in interstadial paleosols of Crvenka (Zech et al., 2013) and Tokaj (Schatz et al., 2011), providing evidence for a rather open N-cycle in interstadial ecosystems as compared to glacial ecosystems. The $\delta^{15}N$ record of Belotinac also indicates an enhanced loss of depleted N-fractions from the interstadial paleosols, as well as for the intercalated loess layers between 2.2 and 6 m depth. Studies on modern ecosystems observed that openness of the N-cycle and pronounced enrichment of soil δ^{15} N can be related to limited water availability and high variability of rainfall (Aranibar et al., 2004; Swap et al., 2004). As N-limited ecosystems efficiently recycle N, the openness of the N-cycle and loss of depleted N is typical for nutrient rich ecosystems (Swap et al., 2004). Hence, high δ^{15} N values in paleosols can be taken as indicators for both semi-arid/arid conditions and higher ecosystem productivity. Semi-arid conditions are indicated by the carbon isotope, macrofossil, and biomarker evidence. As well, the TOC and TN record of Belotinac confirms enhanced ecosystem productivity in the interval from 2.2 to 6 m depth. This is consistent with the results from the <5 μ m fraction proxy of pedogenic clay formation, showing no pronounced changes between the lithological units, but a general slight enrichment in this part of the profile. The formation of loess layers between the interstadial paleosols at Belotinac reflects varying intensity of dust deposition at this site, rather than climatic deterioration.

As the carbon isotope data and other paleoenvironmental proxies indicate humidity as a feature of the glacial units in the Carpathian Basin, the increase of productivity during interstadial conditions, witnessed by the nitrogen isotopic composition, is linked to a more favorable temperature regime. In contrast, the last glacial maximum loess unit L1L1 exhibits lower δ^{15} N values, as well as TOC and TN content, indicating a less productive ecosystem due to cooler climatic conditions. Absolute amounts of precipitation and temperatures as well as duration of the vegetation period likely decreased in the glacial maximum, restricting the overall ecosystem productivity. However, although less biomass was produced in the last glacial ecosystem of the Carpathian Basin, higher levels of soil moisture and the reduced water stress favoured the appearance of conifers.

5. Conclusions

The Belotinac section preserves four main paleoenvironmental phases. The first is related to the deposition of a basal sandy layer associated with a highly dynamic aeolian landscape, indicated by high contributions of coarse sand. The second phase is represented by a succession of MIS 3 paleosols separated by thin loess layers. Paleoenvironmental conditions during this time were cooler. Summer temperatures were lower than today, but not extreme (>12 °C, Sümegi and Krolopp, 2002). Due to reduced temperatures and hence reduced evapotranspiration, soil moisture deficit and dry stress was less intensive than under full interglacial conditions. and ecosystems were relatively productive. Climatic conditions at Belotinac likely were suitable for soil formation throughout MIS 3. Hence, lithological changes between interstadial soils and intermittent loess layers during MIS 3 are more related to changes in the sedimentary system (higher dust input rates) than to any changes of local temperature or precipitation.

During the last glacial maximum (L1L1 loess unit), further cooling and reduction of the potential evapotranspiration increased moisture availability and diminished plant water stress. As summer temperatures were ~>10 °C (the C_4 – C_3 crossover temperature for LGM CO₂ levels), this likely allowed a coeval expansion of tree vegetation and C₄ grasses at Belotinac. Due to lower temperatures (but >12 °C summer temperature; Sümegi and Krolopp, 2002) and a shortened growing season, the overall biomass productivity of this ecosystem was relatively low.

The Belotinac section has great potential to give insight into the paleoenvironmental evolution from a more southern perspective than from the classical loess sites in the Carpathian Basin. This study of the Belotinac section also represents an example for the potential of a dual carbon and nitrogen isotope approach, combined with grain size analyses, for paleoenvironmental reconstruction. Only a small number of similar studies exist for European loess sites. A comparison revealed a zonation from more humidinterglacial and interstadial conditions and drier glacial conditions in the more oceanic-influenced loess area in Eastern France and Western Germany (province of glacial drying), to a loess zone with drier interglacials and increasing humidity in interstadials and even more in glacial periods (province of glacial humidification). The northern part of the Carpathian Basin might represent a transitional area between both loess zones. Further investigations of loess sites, however, are necessary to confirm these results.

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