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# Excursions to C<sub>4</sub> vegetation recorded in the Upper Pleistocene loess of Surduk (Northern Serbia): an organic isotope geochemistry study

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### Abstract

Loess sequences have been intensively studied to characterize past glacial climates of the 40–50  $^{\circ}$  North and South latitude zones. Combining different approaches of sed-imentology, magnetism, geochemistry, geochronology and malacology allows the gen-

- <sup>5</sup> eral pattern of the climate and environment of the last interglacial-glacial cycle in Eurasia and America to be characterized. Previous studies performed in Europe have highlighted the predominance (if not the sole occurrence) of  $C_3$  vegetation. The presence of  $C_3$  plants suggests a regular distribution of precipitation along the year. Therefore, even if the mean annual precipitation remained very low during the most extensive glacial
- <sup>10</sup> times, free water was available for more than 2 months per year. Contrarily, the  $\delta^{13}$ C record of Surduk (Serbia) clearly shows the occurrence and dominance of C<sub>4</sub> plants during at least 4 episodes of the last glacial times at [26.0–28.0], [30.0–31.4], [44.5–53.4] and [66.1–86.8] (in kyrs cal. B.P.). The C<sub>4</sub> plant development is interpreted as a specific atmospheric circulation pattern that induces short and dry summer conditions.
- As possible explanation, we propose that during "C<sub>4</sub> episodes", the Mediterranean Sea would have been under the combined influence of the following: (i) a strong meridional circulation unfavorable to water evaporation that reduced the Mediterranean precipitation on the Balkans; and (ii) a high positive North Atlantic Western Russian (NAWR)-like atmospheric pattern that favored northerlies over westerlies and reduced Atlantic pre-
- <sup>20</sup> cipitation over the Balkans. This configuration would imply very dry summers that did not allow C<sub>3</sub> plants to grow, thus supporting C<sub>4</sub> development. The intra "C<sub>4</sub> episode" periods would have occurred under less drastic oceanic and atmospheric patterns that made the influence of westerlies on the Balkans possible.

### 1 Introduction

<sup>25</sup> Loess deposits are important terrestrial sediment records that provide key data for climate reconstruction and the interpretation of past glacial cycles (Guo et al., 2002;



Kukla, 1977). Combining multidisciplinary approaches (sedimentology, magnetic properties, geochemistry, geophysics, geochronology, malacology) allows a general pattern of climatic and environmental evolution in Eurasia and America to be proposed.

- In Western Europe, high-resolution study of the Nussloch loess sequence (Germany), supported by a large set of luminescence (OSL, IRSL, TL) and <sup>14</sup>C dates, has allowed correlation of the loess grain size variations and loess/paleosol alternation with the Greenland ice-core dust record, which suggests a global connection between North Atlantic and Western European atmospheric circulations and associated wind regimes (Fuchs et al., 2012; Rousseau et al., 2007). The first attempt to model the impact of the abrupt climate variations of the North Atlantic on dust emissions supports the hypothesis that the North-Atlantic millennial-timescale variability is imprinted
- on Western European loess profiles and points to changes of the vegetation cover as the main factor responsible for the dust emissions, yielding material for millennial-scale sedimentation variations (Sima et al., 2009). Among the multidisciplinary investigations,
- <sup>15</sup> a recent organic geochemistry study focused on the impact of these abrupt events in terms of precipitation at the key section of Nussloch. Using inverse modeling of  $\delta^{13}$ C and vegetation, Hatté and Guiot (2005) showed a general glacial precipitation background of 200 mm yr<sup>-1</sup> along the last glaciation punctuated by estimated increases of 100 % recorded during interstadial events.
- A comprehensive pattern of past Western European mid-latitude atmospheric circulation and interconnection is now emerging, but comparatively few similar high-resolution data on past climate are available for Central Europe. Stratigraphical, paleopedological and chronological studies (Antoine et al., 2009a; Fuchs et al., 2008; Galović et al., 2009; Schmidt et al., 2010; Stevens et al., 2011; Zech et al., 2009) in
- Serbia have provided information that the Carpathian region and Western European environments were under different atmospheric conditions that resulted in a drier environment throughout the last climatic cycle (Antoine et al., 2009a; Marković et al., 2008). This conclusion was based on grain-size and paleosol analyses, but a more precise interpretation requires appropriate investigation. Indeed, the extent of this dryness, the



search for seasonality of the precipitation and the reconstruction of past vegetation appear necessary for providing key elements for understanding the past atmospheric circulation conditions in this area.

- Such an issue could be addressed by an organic isotopic geochemistry study, as has already been performed in Western Europe. In contrast to interglacial soils, typical glacial loess is a suitable sediment for organic geochemistry studies, as it accumulates very quickly during the cold oxygen isotope stage (OIS) and is associated with sparse vegetation and a weak rhizosphere. The presence of centimeter-thick laminated structures recognized in most of the typical loess (Derbyshire and Mellors, 1988; Lautridou,
- <sup>10</sup> 1985; Schwan, 1986) implies the absence of significant vertical disturbance and a good preservation of the memory of the climatic conditions contemporaneous to the time of deposition. The lack of conditions favorable to pedogenesis and the dry periglacial environment favor the degradation of organic matter without distortion of the isotopic signal, making typical loess suitable for organic geochemical study (Hatté et al., 1998). The <sup>15</sup> carbon isotopic composition ( $\delta^{13}$ C) of organic matter preserved in typical loess sed-
- iments nicely reflects the original isotopic signature of the vegetation and, therefore, represents an indicator of paleoenvironmental conditions.

The isotopic signature of vegetation provides information on photosynthetic pathways ( $C_3$  versus  $C_4$ ) (Farquhar et al., 1982; O'leary, 1981) and, thus, on environmental changes that are a prelude to the replacement of one vegetation type by another. Based

- <sup>20</sup> changes that are a prelude to the replacement of one vegetation type by another. Based on physiological studies on plants and on the  $C_4$  versus  $C_3$  distribution, a replacement of  $C_3$  by  $C_4$  plants occurs when the  $C_3$  plants can no longer develop because of severe environmental changes, such as changes in altitude, temperature, precipitation and wind along with their seasonal patterns. Ecological niche succession follows the rule of
- <sup>25</sup> "choice of the stronger". If potential niches of  $C_4$  and  $C_3$  plants overlap, the  $C_3$  plants will prevail. Austin (1985) realized that the ecological niche of  $C_4$  plants is the potential niche minus the  $C_3$  overlapping niche.  $C_4$  plants will expand when  $C_3$  plants disappear.  $C_3$  plants need available water for at least 2–3 months, according to the species, to



complete a growth cycle. In contrast, most  $C_4$  plants can complete a growth cycle in less than 2 months with available water (Paruelo and Lauenroth, 1996).

Working at the bulk (plant) scale justifies the use of empirical relationships linking environmental conditions to plant isotopic signatures (concentration and isotopic com-<sup>5</sup> position of atmospheric CO<sub>2</sub>, water availability and, secondarily, temperature, soil type and texture and insolation) previously established at this scale (Lloyd and Farquhar, 1994) and not yet available at the molecular scale.

This study presents new geochemical data obtained from the Surduk loess sequence in Serbia and proposes a new environmental scheme to better understand the past environmental conditions in the south of the Carpathian basin during the last glacial cycle.

### 2 Location, sampling and methodology

### 2.1 Location

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The Surduk loess section is located on the right bank of the Danube River (45°04'N;
 20°20'E, ~ 111 m.a.s.l.) in the southeastern part of the Carpathian Basin ca. 30 km northwest of Belgrade, Serbia (Fig. 1), at the southern edge of the European loess belt.

The area is characterized by the occurrence of thick loess-paleosol sequences that main outcropping in quarries but also as high loess cliffs along the left bank of the Danube River and at the confluence between the Danube and other tributaries, includ-

<sup>20</sup> Danube River and at the confluence between the Danube and other tributaries, including the Tisa River east of the Titel Plateau (Fig. 1). Today, the site is mostly under a Mediterranean climate influence, with winter occurring from November to February. The average annual temperature is 10.9 °C. In January, the average temperature is –1 °C and in July it is 21.6 °C. The annual rainfall is ca. 690 mm, and there are ca. 120 rainy days (Klein Tank et al., 2002).



### 2.2 Sampling

All stratigraphic studies and high-resolution samplings were carried out on a 20-m-high vertical loess cliff over a period of 15 days. Due to stability and security problems, the upper 3 m of the section was sampled in a trench excavated from the top above the vertical profile. The work began with the careful cleaning (removal of weathered material) of the whole section to provide a highly detailed stratigraphical profile (Fig. 1 stratigraphy). This cleaning step is crucial for organic geochemistry to prevent any pollution by organic material, which can be found, according to the sediment texture, as far as 0.5 m below the exposed surfaces. This material can be the product of bacterial activity in the coarser sediment, nets of burrowing insects or the illuviation of organic 10 compounds in topsoil through cracks. Removal from at least 1 m below the vertical wall reduces the contamination risk. Furthermore, measuring the nitrogen content of the sampled sediment checks a posteriori for the absence of modern organic matter. As nitrogen is mostly linked to amino acids that rapidly decrease with organic matter degradation, a measurable level of nitrogen implies the input of recent organic matter 15

in

into the sediment.

The sampling methodology used in Surduk for the geochemistry was based on the continuous column sampling (CCS) method developed by the team several years ago when investigating West European loess sequences. This method consists of cutting a continuous vertical column ( $\pm$ 5–7-cm width) through the whole loess-paleosol sequence, which is then sliced every 5 cm to produce 376 homogeneous samples of sediment. The CCS method allows the geochemistry to be averaged every 5 cm, preventing any gap between the different samples as usually occurs when taking a succession of isolated samples. A single sample was subdivided into four for grain-size, carbon content and  $\delta^{13}$ C and <sup>14</sup>C determination. This division allows the correlation of independent environmental province.

independent environmental proxies. More information on the CCS and on the Surduk sampling is available in Antoine et al. (2009b) and Antoine et al. (2009a), respectively.



Sediment sampling is performed while preventing contact with any organic material, which means no hand contact with the sample at any time and no contact with paper or any potential pollutant, including smoking. Samples are preserved in zipper PE plastic Minigrip bags with no VOC emission. We chose to sample a large amount of sediment (approximately 50 g), even though only some 100 mg is necessary for geo-chemical analysis. This process "dilutes" any potential contamination that would still have subsisted after all the precautions we took. Following this protocol is absolutely necessary for the quality requirement of the investigation of sediment with such a low amount of organic carbon (typically 0.1 %wt) (Gauthier and Hatté, 2008).

### 10 2.3 Geochemistry methodology

The sediment samples were dried at low temperature as soon as possible to ensure safe storage, as recommended by Gauthier and Hatté (2008). After being sieved at 250 µm to remove stones and being homogenized, the sediment then underwent a soft leaching process to remove carbonate using pre-combusted glass beakers, HCl 0.6 N at room temperature, ultra-pure water and drying at 50 °C. The samples were then crushed in a pre-combusted glass mortar for homogenization prior to carbon content and  $\delta^{13}$ C analysis. The handling and chemical procedures are common precautions employed with low-carbon-content sediments.

### Organic and carbonate content

<sup>20</sup> Two different carbon measurements were performed for every sediment sample: total carbon for the bulk sediments and organic carbon for the leached sediments. Approximately 15 to 20 mg of sediment was weighed in tin cups for measurement (with a precision of 1 µg). The sample was combusted in a Fisons Instrument NA 1500 Element Analyzer, and the carbon content determined using the Eager software. A standard was inserted every 10 samples. The inorganic carbon content in the bulk sediment was calculated by assuming that mineral carbon exists only as CaCO<sub>3</sub>. The results



are reported in % weight of carbonate/bulk sediment and in % weight of organic carbon/bulk sediment.

### Carbon isotopic signature

Analysis was performed online using a continuous flow EA-IRMS coupling, that is, a Fisons Instrument NA 1500 Element Analyzer coupled to a ThermoFinigan Delta + XP Isotope-Ratio Mass Spectrometer. Two home internal standards (oxalic acid,  $\delta^{13}C = -19.3\%$  and GCL,  $\delta^{13}C = -26.7\%$ ) were inserted every five samples. Each home standard was regularly checked against international standards. The results are reported in the d notation:

10  $\delta^{13}C = (R_{sample}/R_{standard} - 1) * 1000$ 

where  $R_{\text{sample}}$  and  $R_{\text{standard}}$  are the <sup>13</sup>C/<sup>12</sup>C ratios of the sample and the international standard, Vienna Pee Dee Bee (VPDB), respectively. The measurements were at least triplicated to the representativeness. The external reproducibility of the analysis was better than 0.1 %, typically 0.06 %. Extreme values were checked twice.

### 15 2.4 Geochronology methodology

### **IRSL** dating

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Ten samples were taken for infrared stimulated luminescence dating (IRSL) using cupper cylinders ( $\pm 4$  cm), which were hammered into the loess section to avoid any contamination by light-exposed material. Additional material was taken from the 30-cm surrounding of every IRSL sample for dose rate determination. The sample preparation of the polymineral fine grain fraction (4–11 m), the luminescence measurements and the dose rate determination are explained in detail in Fuchs et al. (2008).



# <sup>14</sup>C<sub>dating</sub>

Based on the δ<sup>13</sup>C results, 15 samples were selected for <sup>14</sup>C dating. The <sup>14</sup>C activity evaluation was performed using AMS physical measurements taken at the Australian ANSTO (ANUA numbers), the NSF-Arizona-AMS-Lab (AA numbers) and the French
LMC14 (SacA numbers) facilities. The CO<sub>2</sub> gas was prepared using three different protocols chosen according to the type of sediment. Hatté et al.'s (2001c) (HCl 0.6 N, Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub> 0.1 M and HCl 1 N at room temperature) was applied for typical loess sediment, whereas either Hatté et al.'s (2001b) (HCl 0.6 N, Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub> 0.1 M, K<sub>2</sub>Cr<sub>4</sub>O<sub>7</sub> 0.1 M/H<sub>2</sub>SO<sub>4</sub> 2 N at room temperature) or Hatté et al.'s (2001c) protocols were applied to sediment extracted from gleys under N<sub>2</sub> flow to avoid possible incorporation of modern CO<sub>2</sub> during alkali treatment by adsorption on Fe<sup>2+</sup>.

All <sup>14</sup>C measurements were converted to calendar ages using Calib 6.0, which includes the IntCal09 calibration (Reimer et al., 2009).

# 3 Results

### 15 3.1 Geochronology

All geochronological data are reported in Table 1 and are shown with their stratigraphic position in Fig. 2. Within errors, the <sup>14</sup>C and IRSL dates are in good agreement. Some classical discrepancies remain only because <sup>14</sup>C and luminescence dating do not characterize the same event. <sup>14</sup>C dating estimates the time elapsed since the death of the plant that trapped the dust, while luminescence estimates the time elapsed since the grains to be dated were without the influence of sunlight. Both chronologies cannot be directly compared, especially for recent times during which external parameters that are at the origin of the discrepancy may be larger than the uncertainties of the physical measurement (Fuchs et al., 2008).



The largest discrepancy between organic radiocarbon and mineral luminescence chronologies occurs between a 4- and 6-m depth (Fig. 2), where the organic chronology has a relatively uniform sedimentation rate. The mineral IRSL would indicate a rupture in the sedimentation at the onset of the major loess accumulation. This discrepancy

- <sup>5</sup> may be the result of the intrinsic nature of both chronologies: vegetation at the origin of the organic matter used for the analysis of the C chronology was present all along this interval, whereas mineral accumulation occurred by pulses (Sima et al., 2009). The organic chronology is thus smoother than the mineral chronology. Nevertheless, the shift is approximately 9 kyrs, and smoothing cannot be the only explanation.
- <sup>10</sup> Although the intent of the chronological framework is to place the organic geochemical signal in time, we privileged the <sup>14</sup>C dating to draw an outline that should encompass the most likely chronological organic framework of the sequence (Fig. 2).

We thus face a very high accumulation during the Middle Pleniglacial with 6 m (from 4.5 to 10.5 m) as an imprint of 10 kyrs (between 27 and 37 kyrs) corresponding to an average sedimentation rate of  $1.7 \text{ mm yr}^{-1}$ . This pattern appears to be unusual, as the highest sedimentation rates are generally observed in European loess during the

Upper Pleniglacial ( $\pm$  OIS 2) and upper Middle Pleniglacial (OIS 3) (Fuchs et al., 2008).

### 3.2 Geochemistry

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All geochemical data are presented in Fig. 3. The organic carbon and carbonate contents are both within the classical ranges observed throughout European loess sequences. These contents respectively vary between 0.2 %wt and 20 %wt, with approximately 4 %wt of organic carbon maximum for modern soil associated to the lowest carbonate content of approximately 8 %wt. The lowest organic content (0.06 %wt) corresponds to the highest carbonate content (40 %wt) during the offset of the penultimate glacial period. Typical values of the last glacial periods are 0.15 %wt and 20 %wt, re-

<sup>25</sup> glacial period. Typical values of the last glacial periods are 0.15 % wt and 20 % spectively.

The  $\delta^{13}$ C signature in Surduk varies from -25.1% for the roots of the modern soil to -22.4% at a 445-cm depth. Such a scheme is outside the current pattern measured in



Western Europe, where isotopic values are always lighter than -23.5%. The heaviest  $\delta^{13}$ C record during the last glacial time in the Nussloch (Germany, Upper Rhine Valley), Villiers-Adam (France, Ile-de-France), Bettencourt-Saint-Ouen and Saint-Saufflieu (France, Picardy) loess sequences are -23.5%, -23.9%, -24.1% and -24.1%, respectively (Hatté, 2000; Hatté et al., 1998).

The isotopic organic record of the Achenheim sequence (France, Alsace) is not considered here, as it was perturbed by both periglacial features and inadequate sample preservation; its highest recorded value was -23.1% (outside the periglacial perturbation) (Hatté et al., 1998). Likewise, we do not consider the -16.9% values obtained by Pustovoytov and Terhorst (2004) in Schattenhausen near Nussloch in some tundra gley horizons, which inexplicably have the lightest  $\delta^{13}$ C in typical loess.

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The Surduk  $\delta^{13}$ C record differs from the other European loess geochemical records not only by the heaviest isotopic episode reaching –22.4‰ at a 445 cm depth (ca. [26.0–28.0 kyrs cal BP) but also by three other episodes of heavy  $\delta^{13}$ C values recorded at 675 cm (–22.8‰ ca. [30.0–31.4 kyr cal BP), 1240 cm (–22.6‰, ca. [44.5– 53.4 kyr cal BP]) and a plateau between 1535 and 1500 cm at –22.85‰ (ca. [66.1– 86.8 kyr cal BP]).

If values lighter than approximately -23.5% were interpreted as exclusively resulting from the degradation of C<sub>3</sub> plants (Hatté et al., 2001a), those of -22.4% to -22.85% likely derive from the degradation of combined C<sub>4</sub> and C<sub>3</sub> plants. Nevertheless, a C<sub>4</sub>/C<sub>3</sub>

- <sup>20</sup> likely derive from the degradation of combined C<sub>4</sub> and C<sub>3</sub> plants. Nevertheless, a C<sub>4</sub>/C<sub>3</sub> plant mixture does not imply that both plants cohabited. Plants with both photosynthetic pathways can have occurred successively during the period represented by the sampling interval, i.e., over ca. 250 yR (in the case of the -22.4% value). As the paleoprecipitation reconstruction by inverse modeling of BIOME4 was only validated for
- $_{25}$  C<sub>3</sub> plants. (Hatté and Guiot, 2005), no quantitative paleoprecipitation can be estimated from the  $\delta^{13}$ C signal.



### 4 Discussion

# 4.1 General last climatic cycle trend

The geochemical records clearly match the classical pattern of the last climatic cycle, with a higher organic carbon content and the lowest  $\delta^{13}$ C during the equivalent to OISs

5, 3 and 1. The carbonate content follows the same pattern, with a lower carbonate content for warmer episodes (OIS 5, 3 and 1) as the result of carbonate leaching during pedogenesis.

According to both the organic chronology and the  $\delta^{13}$ C record, Surduk's last interglacial and early glacial periods cover more than 2 m, from a depth of ca. 1850 to 1600 cm (Fig. 2, units 14 to 12). The Upper Pleniglacial covers the upper part of the

- 1600 cm (Fig. 2, units 14 to 12). The Upper Pleniglacial covers the upper part of the sequence from 825 cm to the upper top, the uppermost meter being crossed by a few deep root tracks down to 200 cm from the Holocene humic topsoil horizon (Fig. 2, units 3 to 1). The boundary between the Lower and Middle Pleniglacial is more difficult to establish. Fuchs et al. (2008) and Antoine et al. (2009a) placed the limit at approximately
- <sup>15</sup> 1300 cm (Fig. 2, boundary between units 10 and 9), whereas the organic record would push the climatic pejoration, the equivalent of OIS 4 (boundary between units 8 and 7), to 1150 cm at the offset of the heaviest  $\delta^{13}$ C values.

Aside from the isotopic excursions toward heavy values, the Surduk loess sequence remains roughly within the same  $\delta^{13}$ C range as other European loess sequences. This

- $_{\rm 20}$  result implies drastic climatic conditions along the last glacial cycle that favored C\_3 plants for most of the time. The current level of precipitation should likely be approximately 200–300 mm yr  $^{-1}$  with respect to other loess sequences, and the C\_3 predominance leads to free meteoritic water distributed along the warm season for most of the last glacial period. The field observation did not provide evidence of a direct effect
- <sup>25</sup> of precipitation on the loess deposits through any drainage characteristics. We must consider that vegetation captured all the precipitation.



### 4.2 Excursions toward C<sub>4</sub> plants and climatic significance

Occurrences of  $C_4$  plants are recorded at [26.0–28.0], [30.0–31.4], [44.5–53.4], and [66.1–86.8] in kyrs cal. BP. Based on physiological studies and on niche theory (Austin, 1985),  $C_4$  plants expand when  $C_3$  plants disappear, i.e., when there are less than 2

- <sup>5</sup> months of available water to allow  $C_3$  plants to achieve a complete growing cycle. Available water means "free" liquid water. Snow and frozen water are not available for plant uptake. The occurrence of  $C_4$  plants during at least 4 episodes during the last glacial in Surduk led to the persistence of climatic conditions that were unfavorable to  $C_3$  development.
- <sup>10</sup> Three potential scenarios can be proposed to describe the climatic conditions relative to the heavy  $\delta^{13}$ C episodes: (i) a short and dry summer with less than 2 months of free meteoritic water during the plant growth cycle; (ii) a snowy summer that does not bring free water that would have been directly assimilated by plants; (iii) temperatures less than 0° C for 8–9 months a year, which would make the permafrost thaw too late
- and the soil too hard to allow C<sub>3</sub> plant roots to penetrate; or (iv) a combination of (iii) with (i) or (ii). In any case, the Surduk results provide evidence of a very strong climatic seasonality that has never been recorded in Western Europe.

Based on the climate reconstructions that derive from European palynological record covering the Last Glacial Maximum, temperatures less than 0°C for 8–9 months are very unlikely, even for anterior periods. Indeed, the summer temperature, even during this extreme time, is 6 to 10°C less than the pre-industrial period (Jost et al., 2005; Leng et al., 2012; Lézine et al., 2010; Peyron et al., 1998). With a reference summer temperature of ca. 20°C (modern summer value), the LGM summer temperature should have been 10 to 14°C. Considering a sinusoidal temperature pattern along the year

with the highest temperatures in summer and the coldest in winter, and even considering a very strong seasonality that would have been represented by a sharp sinusoid, a 10 to 14 °C summer temperature cannot be associated with more than 4–6 months of temperatures under 0 °C. The third hypothesis can thus be ruled out.



Repetitive snowy summers would have been recorded by a specific sedimentological feature (niveo-aeolian laminations), but the feature was not observed here (Antoine et al., 2009a). The second hypothesis can thus be ruled out as well.

- The remaining hypothesis suggests dry (and short) summers for times associated with heavy  $\delta^{13}$ C, which is consistent with malacological studies. To the north (Mišeluk (Marković et al., 2004) and Petrovaradin (Marković et al., 2005)) and south (Ruma (Marković et al., 2006) and Irig (Marković et al., 2007)) of Fruka Gora mountain, we note the virtual absence of the hygrophilous *Succinella oblonga*, which is ubiquitous in the loess north of the Alps, in favor of very abundant steppe taxa, such as *Granaria*
- frumentum, Pupilla triplicata, Chondrula tridens and Helicopsis striata. These taxa are rarely found in Western European loess series (Moine et al., 2005, 2008, 2011) and are more or less frequent in Central Europe north of the Alps (Frank, 2006; Ložek, 1964) and in the Pannonian Basin (Sümegi, 2005), though they are not as common as in the Balkans. Likewise, *n*-alkane investigations performed for the Crvenka loess-paleosol (North Serbia) sequence show that grasses dominated the vegetation cover during the
- (North Serbia) sequence show that grasses dominated the vegetation cover during th whole last glacial cycle (Zech et al., 2009).

Combining the specifications of malacological, organic geochemical and isotopic geochemical investigations yields strong vegetation dynamics during the Middle and Late Pleniglacial, with C<sub>4</sub> episodes highlighted by isotopic geochemistry and short excursions toward mosaic or even forest vegetation elements during C<sub>3</sub> plant periods, as indicated by the sub-domination of forest taxa at Petrovaradin during the Late Pleniglacial (Marković et al., 2005) and a few trees (tall or dwarf) during glacial periods, as indicated by peaks toward high C<sub>31</sub>/C<sub>27</sub> *n*-alkane ratios at Crvenka (Zech et al., 2009). These excursions toward close vegetation cannot be evidenced by isotopic signatures alone that remain within the range of C<sub>3</sub> plants for both C<sub>3</sub> grassland and

signatures alone that remain within the range of  $C_3$  plants for both  $C_3$  grassland and forest.



### 4.3 Possible climatic pattern to explain C<sub>4</sub> episodes

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An explanation of the summer precipitation decline over this part of the eastern Mediterranean basin can be found in both modern meteorological patterns and past climate studies. The Balkan climate is under the combined influence of the Atlantic Ocean and the Mediterranean Sea, as both contribute to regional precipitation. Examples of atmospheric circulation patterns associated with a reduction of precipitation over this part of the eastern Mediterranean basin can be found in both modern and past climate conditions.

Such an example is related to the Heinrich Events (HE). Sierro et al. (2005) showed that HE interrupted the antiphase relationship in deepwater formation between the North Atlantic and Mediterranean because of a large injection of fresh water from melting icebergs at the entrance to the Mediterranean. Lower salinities of Mediterranean surface water resulted in a slowdown of western Mediterranean deepwater overturn, even though cold sea surface temperatures (SSTs) and a drier climate should have

- resulted in enhanced deepwater. A similar but less pronounced pattern of cold SSTs was revealed in the Eastern Mediterranean, where catastrophic arid episodes were connected with Heinrich Events as a result of cold water input in the Eastern Mediterranean Basin, which reduced evaporation and precipitation on the continent (Bartov et al., 2003). The contrast between the strongly reduced SSTs in the western basin and
- the much less reduced SSTs in the Eastern Mediterranean basin was enhanced during the Heinrich Events and favored strong meridional circulation. In the Carpathians, this regime resulted in *less precipitation from the Mediterranean Sea*. The precipitation was even lower for periods that lagged behind the HE or during equivalent Mediterranean meridional circulation-favoring situations.
- Another example related to the Last Glacial Maximum (LGM) can be found based on Alpine evidence and SST reconstructions. Several studies (Florineth and Schlüchter, 2000; Kühlemann et al., 2008, 2009) show that the LGM Mediterranean atmospheric pattern consisted of an amplified meridional winter circulation. This pattern would result



in a northward extension of the Azores High toward Iceland or Greenland, blocking the moisture supply by the westerlies. The situation was further enhanced by expansion and intensification of the Siberian High in winter and spring during glacial times. The most common glacial situation on the Balkans was thus a replacement of the wet west-

- <sup>5</sup> erlies by this blocking situation that was more frequent than that of today. The northward displacement of the polar jet in summer allowed westerlies over Western Europe but less and less precipitation from west to east. This situation resulted in *lower precipitation brought by westerlies* over the Carpathians and even lower precipitation for periods under the influence of an intense Siberian High.
- <sup>10</sup> An explanation for the occurrence of Surduk " $C_4$  episodes" can be proposed by looking at modern meteorological patterns and, more closely, at the patterns that are rarely recorded today but could have occurred during glacial times.

The Mediterranean climate is associated with oscillations in sea level pressure, the well-known North-Atlantic Oscillation (NAO) meridional oscillation, which mostly im-

- pacts the Western part of the Mediterranean basin, and the East Atlantic/West Russia mode (EAWR) that plays a key role in the Eastern Mediterranean precipitation. The EAWR is based on two main anomaly centers that today are located over the Caspian Sea and Western Europe. This mode occurs today from fall to springtime. During the high EAWR periods, northerly winds predominate over the eastern Mediterranean re-
- gion. Positive phases of the pattern are characterized by negative-pressure anomalies throughout western and southwestern Russia and positive-pressure anomalies over northwestern Europe. During the EAWR positive phases, drier than normal conditions are found today in a large eastern region of the Mediterranean Basin (Josey et al., 2011; Krichak and Alpert, 2005). A study by Krichak and Jaspert (2005) clearly showed
- dry and cold northerlies over the Balkans during a high phase (positive EAWR), leading to dry conditions. Transposed to glacial conditions with the Fennoscandian ice sheet covering the north of Europe, such a circulation pattern would bring very cold and dry air masses over the Balkans. This scenario is in agreement with the enhancement in the frequency of storms from the N-NW, as suggested by Antoine et al. (2009b). A high



positive EAWR mode would have resulted in *very cold and very dry summer conditions in the Balkans*.

In the present day climate, a high positive EAWR mode can persist several consecutive months, as happened from the winter of 1992/1993 until May 1993. If, during particular intervals of the glacial period, this mode extended throughout the summer, the result would have been very cold and very dry conditions in the Balkans with a duration long enough to hinder the development of  $C_3$  plants and allow the development of  $C_4$  plants.

Put together, these considerations suggest a climatic schema that fits with the occurrence of the "C<sub>4</sub> episodes". During the four episodes (26.0–28.0, 30.0–31.4, 44.5–53.4 and 66.1–86.8 kyrs cal. B.P.), the Mediterranean Basin was dominated by strong meridional oceanic circulation with low evaporation from the Eastern basin and a high positive EAWR mode reducing the influence of Westerlies and favoring northerlies, both leading to dry and cold summer conditions over the Balkans.

<sup>15</sup> Others periods of the glacial record with  $C_3$  plant dominance would then be associated with lower meridional Mediterranean circulation to a weaker EAWR mode and/or a less intense Siberian High, allowing westerlies to access the Balkans.

### 5 Conclusions

Geochemical records of the Surduk loess sequence show similarities with other European loess sequences. The loess organic matter  $\delta^{13}$ C record evidenced dry and/or cold climatic conditions during glacial times with high  $\delta^{13}$ C values and less drastic conditions during interglacial periods with low  $\delta^{13}$ C. Nevertheless, and in contrast to all European loess sequences recorded along the last climatic cycle, with widespread C<sub>3</sub> plant dominance, the organic  $\delta^{13}$ C record of Surduk is the only glacial record with several unquestionable records of C<sub>4</sub> plants.

This finding suggests a past atmospheric circulation schema over Europe with a focus on Balkan areas. The whole glacial period would be associated with a strong



meridional Mediterranean circulation responsible for a low evaporation rate and with an atmospheric situation unfavorable to the influence of westerlies over the Balkans. This situation would have been enhanced during at least four episodes (26.0–28.0, 30.0-31.4, 44.5-53.4 and 66.1-86.8 kyrs cal. B.P.) under a high positive EAWR-like atmospheric mode that even reduced the Mediterranean evaporation and westerlies in favor of northerlies over the Balkans. This climatic configuration would have led to short and very dry summer conditions unfavorable to  $C_3$  plant development and, therefore, would have allowed the development of  $C_4$  plants.

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**Table 1.** Chronological data of the Surduk loess sequence. The upper panel gathers IRSL dating data (Fuchs et al., 2008). The lower panel is for the <sup>14</sup>C dating. The specificity of the chemical treatment prior to  $CO_2$  evolvement and the <sup>14</sup>C activity measurement is provided in a reference column. The <sup>14</sup>C results are shown as conventional <sup>14</sup>C and calibrated <sup>14</sup>C ages based on the Calib6.0 calibration (Reimer et al., 2009), for which minimum, maximum and median ages are given.

		IRSL age						
depth [m]	sample #	age [yr]	±1σ [yr]					
2.6	BT 140	15.800	1.600					
4.9	BT 141	19.700	2.100					
8	BT 142	36.300	3.900					
8.4	BT 143	31.800	3.400					
9.8	BT 144	39.800	4.500					
11.6	BT 145	53.400	5.600					
12.7	BT 146	53.100	5.500					
14.2	BT 147	66.000	7.000					
15.8	BT 148	82.600	9.000					
19.4	BT 149	120.700	12.800					
				<sup>14</sup> C conv age		calibrated age ±2 $\sigma$ [3]		
depth [m]	chemistry #	physical measurement #	chemical treatment ref.	age [yr]	±1σ [yr]	min [yr]	max [yr]	median [yr]
1.8	GifA-050011	ANUA-31418	[1]	6.400	190	6.855	7.620	7.295
3.3	GifA-070129	AA-78959	[2]	17.135	85	20.060	20.555	20.335
4.45	GifA-070128	AA-78958	[2]	23.740	145	28.005	29.025	28.490
5.3	GifA-050013	ANUA-31419	[1]	26.000	330	30.235	31.195	30.735
6.75	GifA-050014	ANUA-31420	[1]	26.500	370	30.455	31.445	31.040
7.8	GifA-080225	SacA-13476	[3]	26.775	530	30.320	32.195	31.210
8.05	GifA-070127	AA-78957	[2]	27.550	175	31.300	32.175	31.635
8.45	GifA-050015	ANUA-31421	[1]	26.640	340	30.605	31.520	31.135
8.5	GifA-050016	ANUA-31423	[3]	27.870	440	31.310	33.180	32.145
9	GifA-080224	SacA-13475	[3]	28.360	645	31.460	34.245	32.740
9.25	GifA-070126	AA-78956	[2]	28.950	180	33.020	34.460	33.355
9.7	GifA-080223	SacA-13474	[3]	29.335	725	31.875	35.125	33.800
10.25	GifA-080222	SacA-13473	[3]	29.145	710	31.805	34.935	33.630
11.8	GifA-070123	AA-78953	[2]	44.025	1.350	45.230	49.850	47.320

[1]: AAA under air [2]: AAA under N2 [3]: ABOx

9, 187-215, 2013 **Excursions to C**<sub>4</sub> vegetation in Surduk loess C. Hatté et al. **Title Page** Introduction Abstract Conclusions References Figures Tables Back Close Full Screen / Esc Printer-friendly Version Interactive Discussion

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Fig. 1. Location of the Surduk loess sequence. Other series relevant to this study and mentioned in text are also shown.





**Fig. 2.** Stratigraphy and age model of the Surduk loess sequence. Red diamonds are for IRSL dating; the error margin encompasses the 1 sigma variation range (Fuchs et al., 2008). Blue squares represent Calib 6.0 calibrated <sup>14</sup>C dating (Reimer et al., 2009); the error margin encompasses the 2 sigma variation range. The open symbol represents the > 53 000 <sup>14</sup>C conv. year, for which we only have a minimum age. The dotted lines represent an age model envelope that should very likely encompass the chronology of the loess organic accumulation.





**Fig. 3.** Geochemical data of the Surduk loess sequence. The stratigraphical description is from Antoine et al. (2009a). Blue, green, orange and violet curves represent grain sizes greater than 63 m in %, organic carbon content in % wt, carbonate content in % wt and  $\delta^{13}$ C of loess organic matter in ‰ vs. PDB, respectively. All data are presented versus depth. On the right axis, a non-linear time-scale is presented based on IRSL and <sup>14</sup>C dates. Horizontal bars highlight C<sub>4</sub> episodes.





**Fig. 4.** Atmospheric pattern explaining  $C_3$  and  $C_4$  episodes. Upper panel: atmospheric pattern effective during  $C_3$  episodes; Surduk is under a weak but effective influence of westerlies, allowing the more than 2–3 months of available water required for the  $C_3$  growth cycle. Lower panel: atmospheric pattern that prevailed during the  $C_4$  episodes; Surduk is under the strong influence of dry and cold northerlies, leading to less than 3 months of available water. Red arrows are from Kühlemann et al. (2009) and violet arrows are from Krichak et al. (2005) and Josey et al. (2011).

