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Excursions to C4 vegetation recorded in the Upper Pleistocene loess of Surduk (Northern Serbia): an organic isotope geochemistry study
by Hatté C., Gauthier C. , Rousseau D.-D., Antoine P., Fuchs M., Lagroix F., Markovic S., Moine O., Sima A.

To the Editor, Dr Victor Brovkin

Please find bellow extensive replies we made to all three reviewers for the manuscript CP-2012-176. I checked all lines numbers (they refer to lines in the revised manuscript with tracks) and copy-paste on this letter the revisions we made. We thank you for handling this manuscript and hope we have satisfactorily addressed the reviewer's comments.

All the very best

on the behalf of co-authors,
Christine Hatté

legend:
reviewer comment (in blue)
our reply (in black)
=> what we changed in the manuscript (after arrow)

> new/revised text in the manuscript (smaller case and with a border)

Reply to Hong Wang's review

We are grateful for your constructive review, Dr Wang, and feel that your comments are highly valuable contributions to the discussion of our manuscript and help improving our manuscript.

1. phytoliths and their fingerprinting in sediment
we are grateful for this comment that might help to also consider as potential C4 excursions, the events at 8.2-8.3m and 12-12.05m, that occurred about 100-150 years and 300-400 years during the [30.7 - 32.9kyr] and [42.0 - 50.4kyr] intervals respectively.
=> We added a paragraph for these points (§ 3.2, Lines 320-326):

> Furthermore, C4-derived organic carbon decomposes faster than its C3 counterpart in mixed C3/C4 environments (Wynn and Bird, 2007) leading to a shift towards more negative values of the sediment organic d13C by comparison with the plant mixture d13C. This might be of importance in typical loess environment where mineral accumulation rates are high. Therefore, the presence of C4 plants can also be invoked for the events recorded at 825 cm (-23.1‰) and at 1200 cm (-22.9‰) that occurred during the [32.9 - 30.7 cal kyr] and [50.4 - 42.0 cal kyr] intervals respectively

2. C4 dicots and monocots

thank you for this paleoecology point of view that is helpful to explain short episodes as the most recent ones. Annual dicots need less time to expand than perennial plants.

=> We added a paragraph for this point (§ 2.1 Lines 154-157 for modern situation, §3.2 Lines 320-326 and § 4.2 Lines 363-374 for probable past situation).

> The area does not undergo very strong seasonality with dry summer season and/or long and cold winter (Figure 2). This implies a region covered by plants with a carbon C3 fixation pathway. Less than 2% of vascular plants in South-East Europe are C4 plants (Pyankov et al., 2010).

> Furthermore, C4-derived organic carbon decomposes faster than its C3 counterpart in mixed C3/C4 environments (Wynn and Bird, 2007) leading to a shift towards more negative values of the sediment organic $\delta^{13}C$ by comparison with the plant mixture $\delta^{13}C$. This might be of importance in typical loess environment where mineral accumulation rates are high. Therefore, the presence of C4 plants can also be invoked for the events recorded at 825 cm (-23.1‰) and at 1200 cm (-22.9‰) that occurred during the [32.9 - 30.7 cal kyr] and [50.4 - 42.0 cal kyr] intervals respectively.

> Pyankov et al. (2010) explicitly described the C4 taxonomic distribution in Europe and its relation to climatic parameters. They summarized their discussion by stating that "the abundance of total C4 dicotyledons including C4 *Chenopodiaceae* is correlated with precipitation and aridity but not temperature, whereas the abundance of total C4 monocotyledons, C4 *Poaceae* and C4 *Cyperaceae* is correlated with temperature and aridity but not precipitation." Today C4 dicotyledons and C4 *Chenopodiaceae* represent about 65-75% of the C4 plants in the Southeastern and Central Europe, i.e. in the present Surduk geographical region and in the likely modern analog region of past Surduk vegetation. This allows us to consider that C4 dicotyledons and C4 *Chenopodiaceae* were likely the most abundant C4 plants and that their emergence was linked to water availability. So that, C4 plants expand when there are less than 2 months of available water to allow C3 plants to achieve a complete growing cycle.

3. future investigations in loess sites south of the Surduk, Serbia

=> many thanks for your proposition we already followed. Our team was on field in Bulgaria last summer.

4. others

* Figure captions are confused between Figures 3 and 4.

=> we brought back figures and legends between figures 3 and 4.

* In addition, it would be appreciated if keys for stratigraphic units were displayed in either Figure 2 or Figure 4, or both

=> we added keys stratigraphic units in Figures 2 and 3.

* Table 1. For age reporting, a comma "," rather than a period "." should be used.

=> the style of the journal is to use a "space" instead of a comma for large numbers (e.g. 53 000).

* Table 1: It would be appreciated if equivalent dose "De" (Gy), chemistry concentrations derived from ICP-MS or activity ratios for U, Th, and ^{40}K derived from gamma spectrometry, dose rate (Gy/ka), and water contents are included for IRSL age report.

=> We separated both tables in table 1 (for IRSL) and table 2 (for ^{14}C) and we extended table 1 by adding U and Th contents and dose. Further information can be found in Fuchs et al. 2008.

* On page 196, line 6, Hatté et al.'s (2001c) should be Hatté et al. (2001a)?

=> Line 243: Hatté et al. (2001c) should be Hatté et al. (2001b)

* On page 200, line 23, the citation of Lézine et al. 2010 is missing from the References

=> Line 771-776: we added the Lezine et al. 2010 missing reference

> Lézine, A.-M., von Grafenstein, U., Andersen, N., Belmecheri, S., Bordon, A., Caron, B., Cazet, J.-P., Erlernkeuser, H., Fouache, E., Grenier, C., Huntsman-Mapila, P., Hureau-Mazaudier, D., Manelli, D., Mazaud, A., Robert, C., Sulpizio, R., Tiercelin, J.-J., Zanchetta, G., and Zeqollari, Z.: Lake Ohrid, Albania, provides an exceptional multi-proxy record of environmental changes during the last glacial-

interglacial cycle, Palaeogeography, Palaeoclimatology, Palaeoecology, 287, 2010.

* page 203, line 24, assuming it is the typo that "by Krichak and Jaspert (2005). it should be Krichak and Alpert (2005).
=>Line 531: it is effectively Krichak and Alpert (2005).

Reply to Ludwig Zöller's review

We are very grateful for your review, Prof. Zöller, and fully agree that paleoclimatology isotopic geochemistry requires comprehensive explanation, especially to set down fundamentals. We will take benefit of your review to be more accurate and to further develop the pre-requisite and the limitations of the method.

* It is noteworthy that the discussion chapter includes results from disciplines which at the first glance may not appear relevant, e.g. reconstruction of LGM atmospheric circulation based on glacial geomorphology (Florineth & Schlüchter 2000; Kühlemann et al. 2009). Further evidence suitable to support the interpretations of Hatté et al. is given in recent publications by Buggle et al. 2008, 2009 and 2010 which deserve to be mentioned. The publication by Sebe et al. (2011), even if their conclusions about "Mega-Yardangs" in Hungary are still under debate, may also support the proposed model atmospheric pattern explaining C3 and C4 episodes.
=> We thank you to call attention on publications by Buggle et al. 2008, 2009 and 2010 and Sebe et al. (2011). We added these points of view in our discussion: Lines 503-511 and Lines 550-556.

As cold Pleistocene winds move closer to the ground, they are, consequently, more influenced by the topography than during warm periods. The Carpathians can thus deflect original weak westerlies towards N/NW direction (Sebe et al., 2011). This is in agreement with previous investigations performed in the same area. Based on mineral geochemistry investigation on Stari Slankamen loess sequence (Figure 1), Buggle et al. (2008) show that loess originated from alluvial sediments of the Danube and of weathering products of the Carpathian mountain drained by the Tisza and the Drava rivers. They therefore favored a meteorological pattern with strong influence of N/NW winds.

This situation, which predominanted during the last glaciation could also be connected with the N/NW winds indicated by mineral geochemical (Buggle et al., 2008) and sedimentological (Antoine et al., 2009a) tracers as cold Pleistocene winds moved closer to the ground and consequently more influenced by the topography than during warm periods. The Carpathians can thus deflect original westerlies ("wet" winds) towards N/NW direction (Sebe et al., 2011).

* Paleotemperature between LGM and missing modern analogs.
The consideration on palaeotemperatures during the LGM (page 200, last paragraph) is somewhat speculative because modern analogues are missing. Climate stations in Central Asia with summer temperatures up to 14°C and up to 6 months below 0°C lie much farther north and are not summer dry. Presently summer dry steppe climates in Asia lying at lower latitudes have summer temperatures >20°C and very few or none months below 0°. Nevertheless, the consideration is justified but should be classified as somewhat speculative due to the lack of modern analogues

=> We agree that unfortunately modern analogue are missing and that ideal modern analogs don't exist. This equally because such cold temperatures are actually present far north under a quite different diurnal cycle than what can be found at mid latitude in the past. This long daylight during summer (growing season) decreases a little bit the stress induced by the low temperature and won't anyway provide good modern analog of past climate. We added the speculativeness of this part (§4.2, Lines 393-404).

> However, these reconstruction methods were based on assumptions which are not all valid. First, any past pollen assemblage is assumed to be well approximated by the modern analog, but glacial assemblage lack good modern analogues. As example, modern analogues for glacial steppe are missing as they are found today in Central Asia under milder winter and warmer summer. Second, plant-climate interactions are assumed to remain constant throughout time. Implicitly this assumes that these interactions are independent of changes in atmospheric CO_2 and of daylight, whereas a number of physiological and palaeoecological studies (Cowling and Sykes, 1999; Farquhar, 1997; Polley et al., 1993) have shown that plant-climate interactions are sensitive to atmospheric CO_2 concentration and sun exposure. Even considering these restrictions, it is very unlikely that summer temperatures differed by more than 10°C with these reconstructions.

=> Nevertheless by looking at Hatté et al. (2009) we forgot to mention, ecological niche under low CO_2 concentration and at equivalent latitude (run was performed at 47°40'N 6°30'E) for Dwarf Shrub Tundra and Shrub Tundra (expected biomes for the glacial period) yields for mean annual temperature lower by 10-15°C than reference point set at 9.5°C, i.e. MAT of -5 to 0°C. Such a range can not be associated to more than 8 months of temperature lower than 0°C. We also added this reference in the revised manuscript (§4.2, Lines 408-413).

> Furthermore according to Hatté et al. (2009), ecological niche under low CO_2 concentration at equivalent latitude for biomes expected for glacial periods yields mean annual temperatures lowered by 10-15°C with respect to the reference point set at 9.5°C, i.e. mean annual temperatures of -5 to 0°C. Such a range cannot be associated to more than 6 months of temperature lower than 0°C.

* Gocke references

Although details in the methodology of the applied biogeochemistry are beyond my personal expertise I like to encourage the authors to include recent papers by Gocke et al. (2010, 2011, 2012) from the Nussloch site into discussion. According to them, the rhizosphere from interglacial or interstadial tree and shrub vegetation may penetrate the loess several m deep

=> We refer to Gocke study to illustrate the strict difference between typical loess where we authorize paleoclimatic interpretation and paleosol where Gocke et al use to work and where we know and write that only general paleoclimatic trends can be extracted. We already included Zech et al. reference that is more relevant by using equivalent geochemical tools than Gocke et al. but from the paleoclimatological point of view instead of the modern soil science one, taking into account the clear difference between both facies. Modern soils are associated to a more than 15kyrs temperate forest vegetation, no mineral accumulation, temperate humid climate and are the result of strong and efficient pedogenesis that might lead for 2 meters deep, highly organic soils. This is quite different from last glacial conditions with high mineral sedimentation, very weak and short rhizospheric vegetation, drastic cold and dry climate that all didn't provide conditions to develop real pedogenesis and led for few pseudo-gleys of few centimeters (presumably during the most developed interstadials) intercalated between typical loess without any pedogenesis and without any soil. Knowledge from modern soil science can not be directly transposed to the past. To partly illustrate this point, we also added recent papers of Finke and collaborators (Finke et al., 2008, 2012; Yu et al. 2013) who clearly show that conditions to elaborate modern and developed paleosol soils are quite different from what was available during glacial times.

=> We rephrased and added details in introduction (Lines 82-97 and 105-117)

> Such an issue could be addressed by an organic isotopic geochemistry study, as has already been performed in Western Europe if properly conducted. Loess sequence is an alternation of typical loess and paleosols. These two distinct facies must be considered separately as they yield different types of information. European interglacial paleosols are associated to several millennia of temperate forest vegetation, no or very weak mineral accumulation, temperate humid climate and are the result of strong and efficient pedogenesis forming organic soils that can reach up to 2 meters in depth. (Finke, 2012; Finke and Hutson, 2008; Yu et al., 2013). Roots can penetrate the underlying unaltered sediment (Gocke

et al., 2010). By carefully cleaning the vertical wall to remove all potential superficial modern vegetation which can also have laterally penetrating roots, and by conscientiously investigating the sediment to identify and to avoid rhizolith tracks, contamination risks are greatly reduced. Nevertheless by precautionary principle isotopic signal of soils and paleosols (including Bt horizon) and its underlying 1-meter of sediment should be regarded only as support of climatic trends not climatic quantitative information. Conversely typical glacial loess is a suitable sediment for organic geochemistry studies.

Indeed as corroborated by the very low loess organic content, microbial degradation of the weak and low energetic vegetal input in typical loess during glacial times, results in a near-total mineralization of organic matter. This near-complete degradation does not induce isotopic fractionation and the original isotopic signal is preserved. In contrary flourishing vegetation associated to soils and paleosols provide a large amounts of organic matter with a wide range of energetic value. In such an environment, microbes select compounds of high energetic value at the expense of less easily degradable compounds. This results in a selective degradation of organic matter compounds that might bring in isotopic fractionation. In conclusion, the carbon isotopic composition ($\delta^{13}C$) of organic matter preserved in typical loess sediments nicely reflects the original isotopic signature of the vegetation and, therefore, represents an indicator of paleoenvironmental conditions.

* Technical corrections
* Fig. 4; note that Figure Captions are confused between Fig. 3 and Fig. 4!)
=> we brought back figures and legends between figures 3 and 4.

* page 190, line 2: Do not forget to mention palynology
=> we effectively missed palynology as we refered to what was done on Surduk. We added it (Line 50)

* page 191, line 25: Is it “the stronger” or “the better adopted”?
=> it is the "stronger". both can be adapted but the one who can deliver the highest NPP is the one that will take the place.
* page 192, line 19: page 192, line 19: “main” appears to be wrong. “are outcropping”?
=> "... that main outcropping ..." is replaced by "... that mainly outcrop ..." (Line 148)

* page 192, line 20: remove “other”
=> "other" is removed (Line 149)

* page 195, line 17: “copper” instead of “cupper”;
=> "copper" is replaced by "cupper" (Line 230)

* page 195, line 18: 4 cm diameter (see Fuchs et al. 2008).
=> "(± 4 cm)" is replaced by "(± 4 cm diameter)" (Line 130)

* page 197, line 9: Can you give another possible explanation, e.g., IRSL underestimate of sample BT141 for reasons so far unknown?
=> we added the reviewer proposed explanation of the 9kyr shift between organic and OSL chronologies. (Lines 270-271)

* page 198, line 17: For this period (ca. 66.1 to 86.8 kyr) no 14C-ages are available, only IRSL ages. Therefore, the notation cal BP is not appropriate. Quote as "kyr old (IRSL)". This concerns also other parts of the text further below.
=> we removed "cal" for all dates older than 50kyr if individually presented (i.e. if only one unit is used for several ranges we kept cal. yr BP as 3 out of the 4 ranges are within the ^{14}C ranges. That is the case of 1 occurrence within the text: line 308). Other mention is associated to intervals 14C-datables and we thus left "cal".

* page 199, lines 21 and 22: What is meant by “should likely be approximately 200-300 mm/y”? The nearby Belgrade station actually has 690 mm/y!

=> "current" is the wrong word, it should have been "typical". We corrected it.

Reply to Guo Zhengtang's review

We would like to express our sincerest thanks for your review, Prof. Guo, for your highly constructive comments and suggestions, which are very useful for improving the paper.

* It might be finer to add a figure showing the modern seasonal distributions of precipitation and temperature in the study region because the discussion on the paleoseasonality is a strong aspect of this study

=> we added it as figure 2 as illustration of the modern situation in § 3.2. We numbered other figures in accordance (we also noted we missed to refer to Figure 5, former figure 4, we corrected it).
In order to illustrate the seasonality, we chose to show the annual distribution of precipitation.

* It would be more interesting to add a paragraph addressing the role (significant or not) of temperature on the occurrence of the C4-dominance.

=> We added a mention to the modern plant distribution in the Surduk (§2.1 Lines 155-157)

> This implies a region covered by plants with a carbon C3 fixation pathway. Less than 2% of vascular plants in South-East Europe are C4 plants (Pyankov et al., 2010).

=> and refer to Pyankov et al. (2010) study (as suggested by Hong Wang) to explain the past C4 plants taxonomic distribution (§4.2 Lines 363-375). We likely faced C4 plants (dicots and chenops) that mostly react to precipitation.

> Pyankov et al. (2010) explicitly described the C4 taxonomic distribution in Europe and its relation to climatic parameters. They summarized their discussion by stating that “the abundance of total C4 dicotyledons including C4 *Chenopodiaceae* is correlated with precipitation and aridity but not temperature, whereas the abundance of total C4 monocotyledons, C4 *Poaceae* and C4 *Cyperaceae* is correlated with temperature and aridity but not precipitation." Today C4 dicotyledons and C4 *Chenopodiaceae* represent about 65-75% of the C4 plants in the Southeastern and Central Europe, i.e. in the present Surduk geographical region and in the likely modern analog region of past Surduk vegetation. This allows us to consider that C4 dicotyledons and C4 *Chenopodiaceae* were likely the most abundant C4 plants and that their emergence was linked to water availability. So that, C4 plants expand when there are less than 2 months of available water to allow C3 plants to achieve a complete growing cycle.

* P201 mentions some published malacology data at/near the site. Adding one or two curves of the malacology assemblages in Figure 4 would help the discussions about the environmental conditions.

it would make sense indeed to compare d13C directly with malacological record obtained on the same sequence or at least on a sequence with a similar temporal resolution. Unfortunately Surduk malacological investigation is still in progress and data available for surrounding sequences were not sampled for the same scientific issue. Their aim was more to get a wide (spatially and temporally) overview of the malacological distribution and of the derived paleo-ecological interpretation than to focus at high resolution on one climatic cycle or on some events as we did here. Thanks to these previous studies, we knew that this place was of great interest for past atmospheric circulation reconstruction Temporal resolution of previous malacological studies and sequence thicknesses are definitely lower than the d13C study performed on Surduk. As C4 episodes are tiny and recorded by less than 30cm of loess, they are included into the larger malaco samples extracted from surrounding sequences. Because of this large difference of temporal resolution it is not possible to "correlate" Surduk loess sequence with surrounding ones as precisely as this study requires. By looking at malacological assemblages we found occurrences of some species that fit with our results but we don't know exactly their timing.

=> We are not able to accede reviewer request. Anyway we dug out into archives and literature and completed a little bit this part (Lines 427-436)

> In Ćirikovac and Klenovnik, about 80 km south-east of Surduk, on the western flank of a north-south elongated relief, similar general observations have been recorded with some differences. *S. oblonga* is poorly represented in Ćirikovac, and among steppe taxa only *C. tridens* and *G. frumentum* are abundant, *P. triplicata* and *H. striata* being absent (Mitrović, 2007). However, we must keep in mind that only a single taxa has been sampled in these last two sites. Other identified species suggest a resemblance with more humid and woody steppe vegetation from Ruma and Irig north of the Fruska Gora mountain. Furthermore, fauna from Požarevac brickyard, a few kilometers north of Ćirikovac, indicates even drier environment than in Irig for example (Jovanović, 2005; Jovanović et al., 2006).

Other changes

=> we rephrased our reference to Zech et al. (2009) study to be more precise (Lines 437-453)

> *n*-Alkane investigations performed for the Crvenka loess-paleosol (North Serbia) sequence show that grasses dominated the vegetation cover during the whole last glacial cycle (Zech et al., 2009). However Zech et al. (2009) underlined several periods with presence of trees based on corrected n-alkane distribution. The applied correction derives from modern observation of n-alkane distribution in vegetation and in the associated litter and topsoil where they evidenced a modification of the original vegetation n-alkane distribution in litter consecutively to degradation that conceals the trees percentage in the original vegetation ratio. Middle paleosol complex likely has undergone similar degradation effect but the corrected ratio of trees in typical loess may be overestimated as vegetal organic matter degradation was quite different during glacial times. It is conceivable that as a result of the very drastic conditions and of the weak vegetal input, the original n-alkane distribution was better preserved in typical loess than in middle paleosol (high over-even-odd predominance stated by authors in typical loess, i.e. L1Lx units) and thus loess n-alkane distribution does not require high correction. This said, the possible occurrence of some trees in protected areas during the C3 plants interval remains. Few dwarf trees in open grassland, as currently found today in Greenland, may have grown in Surduk area throughout these periods.