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1. Introduction

 Loess deposits preserved the 'fingerprints' of centennial and millennial scale climatic and environmental changes (Porter and An, 1995; Rousseau et al., 2007; Stevens et al., 2007, 2008; Sun et al., 2012). However, accurate and high-precision chronologies are required to fully utilize and exploit the potential of these key terrestrial archives in reconstructing millennial scale climatic events and understanding their causes and regional consequences. Major obstacles of establishing such loess chronologies are the inherent limitations of luminescence techniques (relatively low precision of OSL/IRSL ages) and often the lack of reliable target materials for radiocarbon dating (e.g. Pigati et al., 2013; Kadereit et al., 2013; Újvári et al., 2014). While charcoal is commonly regarded as a material that can yield reliable 14 C ages (Trumbore, 2000; Hatté et al., 2001b), other materials often yield either anomalous ages (rhizolites; Pustovoytov and Terhorst, 2004; Gocke et al., 2011; Újvári et al., 2014), or dates that are compromised by secondary overprinting (organic matter; Gocke et al., 2010, 2011) or vertical migration of organic contaminants in the soil/loess profile (humic substances; McGeehin et al., 2001; Ascough et al., 2011a; Wild et al., 2013). Unfortunately, 49 charcoal is not abundant in loess deposits and so we must turn to alternative materials for ^{14}C - dating. Mollusc shells are often found in loess sediments (Sümegi and Krolopp, 2002; Moine et al., 2008) and may be useful for establishing firm chronologies. However, some basic 52 questions must be addressed before establishing charcoal and mollusc-based 14 C-chronologies 53 of loess records: 1) how reliable and reproducible are the ${}^{14}C$ ages of charcoals in loess, 2) do some of the mollusc species provide more accurate ages than others, i.e. which species are preferred for 14 C-dating of loess, and 3) what are the characteristic uncertainties of Bayesian 56 age-depth models resting on charcoal/mollusc ${}^{14}C$ dates? In this study we present an extensive dataset of 64 radiocarbon ages obtained by AMS 14 C-

dating of charcoal fragments and shells of various small (<10 mm) mollusc species. These

 materials were recovered from loess sediments collected in the Dunaszekcső sequence in Hungary. A few of these data have already been published in a companion paper discussing 14° C and OSL/IRSL dating of the studied loess record (Újvári et al., 2014), but here we present 62 a more comprehensive ${}^{14}C$ dataset. Using different protocols of charcoal combustion we demonstrate that the applied methodology can sometimes have a profound effect on the 64 resulting charcoal ${}^{14}C$ ages, with differences between measurements ranging from some hundreds of years up to one/two thousands of years. By comparing the charcoal ages with those obtained from mollusc shells we show that loess records can be dated with a relatively good precision using small molluscs. Although some species such as Succineidae sp. (most probably *Succinella oblonga)*, *C. tridens*, *T. hispidus*, and the members of the Clausiliidae family show good potential for dating, the spread of reference charcoal ages obtained from a loess layer containing both charcoals and molluscs prevent us from assigning those mollusc 71 species which are the most preferable to use in ${}^{14}C$ -dating of loess. This implies that further work is needed on the subject, and that, at least at the present state of knowledge, the best dating strategy is to use multiple mollusc species from each sampled stratigraphic interval to establish loess chronologies.

2. Materials and methods

2.1. Study site and sampling methods

 The studied loess-paleosol section is located at Dunaszekcső, Southern Hungary (46°05'25"N, 18°45'45"E, 135 m a.s.l.; Fig. 1) and exposes glacial-interglacial sediments with a thickness of 17 m. After cleaning of the sediment surface, altogether 31 loess samples were collected 81 for radiocarbon dating throughout the profile at various depths from 1010 to 250 cm. Loess 82 cuboids with dimensions of $15 \times 5 \times 10$ cm (length-width-height) at depths of 400, 500, 600 83 cm, $15 \times 5 \times 7.5$ cm at depths of 820 and 825 cm and $15 \times 5 \times 5$ cm at other depths were cut

 from the uppermost loess unit. Sample blocks were subsequently disintegrated in the lab by soaking them in distilled water. Charcoals and gastropod shells (Fig. 2) were extracted by 86 washing the sediments through a 1 mm mesh sieve then dried at 50 \degree C and handpicked using gloves and pre-cleaned forceps to avoid modern carbon contamination. After being identified at the species (or family) level, shells were wrapped in Al-foil and put in closed plastic bags. Also the charcoal samples were handled and packed in a similar way, but separately from land snail shells. The nomenclature of mollusc species follows Welter-Schultes (2012).

2.2. Radiocarbon dating

 Gastropod shells and charcoals were further pretreated in the AMS laboratory of the Hertelendi Laboratory of Environmental Studies, Institute for Nuclear Research, Debrecen, Hungary (Molnár et al., 2013a). During this procedure, charcoal fragments were treated using the standard acid-base-acid (ABA) method (Jull et al., 2006), i.e. in a sequence of 1N HCl, rinsing in distilled water, 1M NaOH, rinsing again in distilled water, and then 1N HCl at 75 ºC, for 1-2 hours each step. After the final acid wash, the samples were washed again with 99 distilled water to neutral pH $(4–5)$ and dried at 60 °C. During the first set of experiments, dried charcoal fragments were combusted in an on-line combustion system using CuO in one 101 step at $1000 \, \text{°C}$ (ABA-OSC₁₀₀₀), whereas during subsequent runs they were subjected to 102 stepped combustion in pure O_2 gas atmosphere, first at 400 °C, and then at 800 °C (ABA- $TSC₄₀₀$ and ABA-TSC₈₀₀). Mollusc shells were ultrasonically washed and then all the surficial contaminations and

 carbonate mineral coatings were etched using weak acid (2% HCl). This etching effectively removed 20‒30% of the shell material. Subsequently, acid cleaned shells were dried and put 107 into vacuum tight two finger reaction ampoules $({\sim}100 \text{ cm}^3$ inner volume) and dissolved by

108 phosphoric acid. $CO₂$ was produced by acidic hydrolysis of shells, further purified

cryogenically and then graphitized (Molnár et al., 2013a).

110 All the 14 C measurements were done on the graphitized samples using a compact radiocarbon AMS system (MICADAS) developed at the Paul Scherrer Institute and the ETH Zürich (Synal et al., 2007; Wacker et al., 2010), which was installed at the Hertelendi Laboratory of Environmental Studies, Debrecen in 2011 (Molnár et al., 2013b). Conventional radiocarbon ages were converted to calendar ages using OxCal online (version 4.2; Bronk Ramsey, 2009) and the IntCal13 calibration curve (Reimer et al., 2013).

2.3. Age-depth modeling

 Bayesian age modeling has been performed using Bacon (Blaauw and Christen, 2011), based on 56 radiocarbon data out of 57 available from profile 1 of the studied loess record between 8.65 and 2.50 m (Table 1). Combined with an estimated starting date for the first section, accumulation rates are estimated from 27.7 million of Markov Chain Monte Carlo (MCMC) iterations and then these rates form the age-depth model. Accumulation rates in this case were constrained by the following prior information: 'acc.shape'=1.5 and 'acc.mean'=10' for the gamma distribution describing accumulation rates, and 'mem.mean'=0.7 and 'mem.strength'=4 for the beta distribution describing the memory effects of accumulation 126 rates. All input age data were provided as ${}^{14}C$ yr BP and Bacon used the IntCal13 calibration curve to convert conventional radiocarbon ages to calendar ages. Age modeling was run to achieve 5 cm final resolution.

3. Results and discussion

 3.1. Charcoal age accuracy and reproducibility: the effects of applied combustion protocols Charcoal is produced during pyrolysis accompanying the incomplete combustion of woody plant tissue under conditions of restricted oxygen (Bird, 2006; Braadbart and Poole, 2008; Bird and Ascough, 2012). A characteristic feature of the resulting fire-altered biomass is the presence of chemically stable aromatic ring structures formed by lignocellulose degradation (Schmidt and Noack, 2000; Preston and Schmidt, 2006). Aromatic rings increasingly form ordered microcrystalline domains with increasing pyrolysis temperatures (Bird and Ascough, 2012), leading to higher chemical stability (Goldberg, 1985). Although charcoals are regarded as relatively resistant to post-depositional alteration (Schmidt and Noack, 2000), there is a growing body of evidence of charcoal degradation and loss by chemical oxidation (Cohen-Ofri et al., 2006; Ascough et al., 2011a,b), physical fragmentation (Gavin, 2003), or fungal degradation (Ascough et al., 2010). Charcoal alteration is mainly controlled by production variables (heat temperature, heating rate, wood properties; Braadbart and Poole, 2008) and the chemistry, especially the pH of the depositional environment (Braadbart et al., 2009). Also, charcoal can readily adsorb a range of soluble chemical contaminants migrating in the sediment column like humic substances, 150 which can have a different ${}^{14}C$ age than the charcoal (Alon et al., 2002; Rebollo et al., 2008; Wild et al., 2013). Obviously, this exogenous carbon must be removed prior to radiocarbon dating, which is commonly done by treating the samples with a series of weak acid and base washes (acid-base-acid=ABA treatment; de Vries and Barendsen, 1954; Olson and Broecker, 1958). The initial acid wash with HCl is intended to remove soluble carbonates and other minerals, while the subsequent alkali treatment (NaOH) removes humic substances (Alon et 156 al., 2002; Rebollo et al., 2008). The final acid wash is designed to remove atmospheric $CO₂$ incorporated into the organic structure during the alkali/base step (Head et al., 1996; Hatté et

 al., 2001a). While the ABA-technique appears to be a robust method for contaminant removal for a number of samples (Rebollo et al., 2011; Bird and Ascough, 2012), several studies demonstrated that it does not always remove all contaminant carbon, which becomes critical 161 with increasing sample ${}^{14}C$ age (Gillespie et al., 1992; Chappell et al., 1996; Douka et al., 2010; Southon and Magana, 2010; Wood et al., 2012). An alternative pre-treatment technique, called the ABOX-SC method, involves an oxidation step after the acid-base steps, which is followed by stepped combustions at 330, 630 and 850 ºC to remove any final traces of labile carbon (Bird et al., 1999). Although this technique proved to be very effective in removing contamination from old samples (Douka et al, 2010; Wood et al., 2012; Bird et al., 2014), it often leads to large losses in sample material (Bird and 168 Ascough, 2012). Therefore, to test previously published ABA-OSC₁₀₀₀¹⁴C ages of charcoals 169 used as reference ages in comparison with mollusc shell ^{14}C ages (Újvári et al., 2014), 170 radiocarbon ages were obtained in a two-step combustion (400 and 800 °C) procedure after ABA-treatment in this study. This method was applied on relatively well-preserved charcoals from depths of 8.25 and 8.20 m (Dsz-Ch1 and 2, profile 1; Table 1; Fig. 3), and poorly preserved older charcoal fragments from a depth range of 10.10 to 8.50 m (profile 2, Table 1; Fig. 4).

175 In a previous study by Újvári et al. (2014) two ABA-OSC₁₀₀₀ ages of $25,568\pm105$ and 176 26,101 \pm 110¹⁴C yr BP were reported from samples Dsz-Ch1 and 2. Since the publication of these ages, the AMS Lab at Debrecen has done extensive chemical blank correction studies using fossil wood samples of known isotopic compositions to better estimate contamination introduced during the chemical preparation of charcoals. These test results indicated that a higher chemistry blank correction is more realistic and has to be applied on the ages published 181 in 2014. Using the new correction factor the above-mentioned $ABA-OSC₁₀₀₀$ charcoal ages 182 were recalculated and are reported here as $25,868\pm165$ and $26,433\pm178$ ¹⁴C yr BP (DeA-2917

 and DeA-2923; Table 1). In comparison to these ages, another charcoal fragment from the 184 same horizon (Dsz-Ch1) yields slightly older ABA-TSC₄₀₀ and TSC₈₀₀ ages (26,726 \pm 142 and $27,320\pm158$ ¹⁴C yr BP; Table 1). The age discrepancy between ABA-OSC₁₀₀₀ and ABA-186 TSC_{400/800} ages from the sampled stratigraphic interval (Dsz-Ch1/2) may partly be attributed to real age differences between the charcoals as the individual fragments dated during the two runs were different and were recovered from a 7.5 cm thick sediment layer integrating some hundreds of years of loess deposition. At the same time, it is obvious that some exogenous carbon contamination remained fixed in the charcoal structure after ABA-treatment as 191 demonstrated by the -590 \pm 220¹⁴C yr difference (=~0.26 pMC) between the 400 and 800 °C fractions. Since the labile carbon was not removed by an additional lower temperature combustion step in the first run, the possibility that those ages are slight underestimates of the true ages cannot be excluded. Two-step combustion ages of coeval and older charcoals from profile 2 further corroborate the above observation of remaining contaminants after ABA- treatment. For samples Dsz/2sz-RC22 and 26, the age differences are -1350±250 and - \pm 780 ¹⁴C yrs (~0.59 and 0.48 pMC), with the ABA-TSC₈₀₀ ages being consistently older 198 than the ABA-TSC₄₀₀ ages (Table 1; Fig. 4). All these data highlight that ABA-TSC₈₀₀ ages give, or at least are closer to, the real ages of charcoals.

 3.2. Charcoal versus shell ages of small molluscs: implications for the 'Limestone problem' and open-system behavior

Early studies documented that land snail shells yield anomalously old radiocarbon ages

204 reaching 3000 yrs offset due to incorporation of old, ${}^{14}C$ -free carbonate from the local

substrate into shell carbonate. This phenomenon is often quoted as the 'Limestone problem'

(Rubin et al., 1963; Tamers, 1970; Evin et al., 1980; Goodfriend and Hood, 1983; Goodfriend

and Stipp, 1983; Yates, 1986; Goodfriend, 1987; Zhou et al., 1999; Xu et al., 2011). However,

208 most of these works were biased towards gastropods having relatively large shells (>20 mm) 209 and recent studies by Brennan and Quade (1997) and Pigati et al. (2004, 2010, 2013) 210 demonstrated that reliable ${}^{14}C$ ages can be obtained from smaller gastropods (shells <10 mm) 211 that have largely been ignored in previous ${}^{14}C$ -dating studies. Beyond the 'Limestone' 212 problem', another one is to assess whether the shells behaved as close systems with respect to 213 carbon during burial. Open-system behavior is a serious concern in older samples (25–60 ka) 214 where small amounts of contamination cause large bias/errors in ${}^{14}C$ ages. Rech et al. (2011) 215 and Pigati et al. (2010, 2013) revealed, by measuring the 14 C-activities of very old mollusc 216 shells (800–130 ka) and testing land snail shell ages against plant macrofossil ^{14}C ages, that 217 many fossil gastropod shells do not suffer from major (>1%) open-system problems. 218 Ten different mollusc species and two additional ones that could be identified only at the 219 family level have been radiocarbon dated in this study, some of them are displayed in Fig. 2. 220 Out of the eleven, shells of 7 mollusc species could directly be compared to reference 221 charcoal ages from samples Dsz-Ch1/2. These shell ages range from 15,844±56 (*Trochulus hispidus*) to 26,979 \pm 126¹⁴C yr BP (Clausiliidae sp.; Table 1). The apparently young ¹⁴C age 223 of *T. hispidus* (15,844 \pm 56¹⁴C yr BP) is attributed to open-system behavior. 224 As shown in Fig. 3, all of the land snail shell ages obtained in the first run overlap within 2σ 225 errors with the ABA-OSC₁₀₀₀ charcoal ages from the same sample except for the anomalous 226 age (DeA-2918) mentioned above. Out of the analyzed shells that of Succineidae sp. show 227 one of the lowest age anomalies $(-290\pm 220^{14}C)$ yr; Dsz-Ch1; Table 1) compared to the 228 conventional age of charcoals obtained by the ABA-OSC₁₀₀₀ protocol (Table 1; Fig. 3). At 229 first glance, this observation would confirm previous findings of Pigati et al. (2010, 2013) that 230 the genus *Succinella* may yield reliable ${}^{14}C$ ages. However, the Succineidae sp. age is clearly 231 younger than the ABA-TSC $_{800}$ age (second run; Table 1), which is considered to be closer to 232 the real age of sedimentation. The negative age anomalies, ranging from -290 ± 220 to -

257 be appropriate targets for ${}^{14}C$ -dating of loess, but the above findings should be further investigated in subsequent studies of other loess sites where charcoals are available. A comparison of Succineidae sp. ages with those obtained from shells of *E. fulvus* and *O. dolium* recovered from 2 samples between 6.25 and 5.75 m demonstrates that the latter two 261 display minor age differences (100 ± 180 and -140 ± 160 ¹⁴C yrs) compared to Succineidae sp. (Dsz-RC12, 13; anomalies not tabulated in Table 1). While no independent data on the use of *O. dolium* for dating late Quaternary sediments exist, Pigati et al. (2004, 2010) found that 264 shells of live *E. fulvus* show ¹⁴C activities that are indistinguishable from live plants and display negligible 'Limestone effect'. If their results could be applied to the fossil record, should be the subject of further testing, as the data presented here are somewhat ambiguous for *E. fulvus* (see Fig. 3, second run).

 3.3. Bayesian age-depth model of the studied loess record and associated chronological uncertainties

271 Bayesian age-depth modeling was performed using 56 AMS 14 C dates for profile 1 from 8.65 to 2.50 m. Two ages were omitted in age modeling as they were clear outliers (Table 1), while no preference or extra weight was given to any age data obtained from particular species 274 identified as promising targets for AMS 14 C dating in comparison with charcoal ages above. This is because of the scatter in charcoal age data in samples Dsz-Ch1/2 (Fig. 3) that rendered the comparison of charcoal and shell ages somewhat ambiguous. Also, it is believed that such a Bayesian age model includes the treatment of the stochastic nature of how the passing of time (as loess sedimentation proceeds) is recorded in shells of small molluscs. Random, hence unpredictable elements of this process (among others) are the minimal, but in some cases still acting 'Limestone effect' of different magnitudes and the possible minor open-system behavior. It is believed that the Monte Carlo-based age model emerges from this noise

 imposed on the true age of the sequence, and if a large number of absolute dates are available both from each dated layer and at many depths, the model will represent the real age of loess sedimentation which is an inhomogeneous Poisson process.

 The software 'Bacon' divided the sequence into 124 vertical sections and accumulation rates were estimated through 27.7 million MCMC iterations, which resulted in an age-depth model 287 presented in Fig. 5. Two post-IR IRSL $_{225}$ ages published earlier (Újvári et al., 2014) are also shown in Fig. 5 to further confirm the validity of the age model. However, OSL and post-IR 289 IRSL₂₉₀ ages (not shown in Fig. 5) are consistently older as discussed in Újvári et al. (2014). The Bayesian age model has a mean 95% confidence range of 464 yr. While the minimum 2σ uncertainty is as low as 238 yr at 4.90 m, the maximum reaches 804 yr at 7.50 m (Fig. 5). Broadening of the 95% confidence interval at around a depth of 7.50 m is due to the lack of

293 14 C ages from 7.75 to 7.15 m in the record (Bennett, 1994).

5. Conclusions

 Shells of various mollusc species and charcoal fragments at multiple depths from the 297 Dunaszekcső loess sequence, Hungary were dated by ${}^{14}C$. Charcoal ${}^{14}C$ ages were found to be 298 dependent on the applied protocol used, with the $ABA-TSC_{800}$ protocol giving consistently 299 the oldest ages. Since the combustion step at 400 $^{\circ}$ C is thought to remove labile, exogenous carbon still present in the charcoal structure after the ABA-treatment, the ages obtained in the 301 800 °C combustion step are considered to be the closest to the real age of the sediments. Comparison of charcoal ages with those of land snail shells reveals that some small (<10 mm) molluscs (Succineidae sp., *C. tridens*, *T. hispidus* and members of the Clausiliidae family) 304 appear to yield accurate, reliable ${}^{14}C$ ages, although the comparison is somewhat uncertain considering the scatter in charcoal ages. Unfortunately, this prevents us from identifying the best mollusc targets for ¹⁴C dating of loess records. Other species, including *E. fulvus*, *D.*

 ruderatus and *O. dolium*, may be useful, but further work involving these species is needed to validate this and the above observations. As the Bayesian model for the Dunaszekcső loess record demonstrates, relatively good chronologies can be gained by dating multiple species 310 from depth intervals of 20-30 cm. The resulting age models will likely have 2σ uncertainties on the order of ca. 500-800 yr.

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Figure captions

- **Figure 1.** Location of the Dunaszekcső loess sequence in the Carpathian Basin.
- **Figure 2.** Some mollusc species included in this study. a) Clausiliidae sp. (sample Dsz-Ch1),
- b) *Cochlicopa lubrica* (Dsz-RC20), c) *Discus ruderatus* (Dsz-RC7), d) *Euconulus fulvus*
- (Dsz-RC8), e) *Nesovitrea hammonis* (Dsz-RC9), f) *Orcula dolium* (Dsz-RC3), g) Succineidae
- sp. (most probably *Succinella oblonga*) (Dsz-RC12), h) *Trochulus hispidus* (Dsz-1R), i)
- *Vallonia costata* (Dsz-RC6), j) *Vitrea crystallina* (Dsz-RC4). Scale bars represent 2.5 mm.
- **Figure 3.** Comparison of conventional AMS radiocarbon ages of charcoal fragments and
- mollusc shells obtained during the first and second runs from samples Dsz-Ch1 and 2 (depths
- 8.20, and 8.25 m). Note that the youngest, obviously anomalous shell age (DeA-2918) from
- sample Dsz-Ch1 is not displayed in this figure.
- **Figure 4.** Comparison of conventional AMS radiocarbon ages of charcoal fragments between
- 521 depths of 10.10 to 8.50 m. Note that $ABA-OSC_{1000}$ ages are from profile 1 (first run), while
- 522 ABA-TSC_{400/800} ages are from profile 2 (second run).
- **Figure 5.** Bayesian age-depth model for the 8.65-2.50 m part of the Dunaszekcső loess record
- 524 and calibrated mollusc shell and charcoal ¹⁴C ages (mean age $\pm 2\sigma$). Two post-IR IRSL₂₂₅
- ages are also shown for comparison (errors are 2σ; data from Újvári et al., 2014). Parameters
- 526 of the age-depth model as described in the 'Materials and methods' section. Note that ${}^{14}C$
- ages from profile 2 were excluded from modeling.

Figure1_color[Click here to download high resolution image](http://ees.elsevier.com/quageo/download.aspx?id=76328&guid=08166c60-2291-4e0d-968a-5bb48606b66e&scheme=1)

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Radiocarbon age ± 20 error

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Table 1. AMS ¹⁴C data of charcoal fragments and mollusc shells from the Dunaszekcső loess record

Profile 2

All the ¹⁴C dates are calibrated by OxCAL Online (version 4.2) using the IntCal13 calibration curve
"Calculated as ABA-OSC₁₀₀₀ – ABA-TSC₈₀₀ or ABA-TSC₄₀₀ – ABA-TSC₈₀₀, rounded to the closest ten (or up in case of

$$
\sqrt{\sigma_{ABA-OSC1000\;or\;ABA-TSC400}^2 + \sigma_{ABA-TSC800}^2}
$$

 $^{\text{b}}$ Calculated as $^{\text{14}}$ C age_{shell} - $^{\text{14}}$ C age_{charcoal (ABA-OSC1000)}, rounded to the closest ten (or up in case of errors), errors propagated as

$$
\sqrt{\sigma_{14C-age-shell}^2 + \sigma_{14C-age-charcoal}^2}
$$

^cCalculated as ¹⁴C age_{shell} - ¹⁴C age_{charcoal (ABA-TSC800)}, rounded to the closest ten (or up in case of errors), errors propagated as in
b)

For definitions of the ABA-OSC₁₀₀₀ and ABA-TSC_{400/800} protocols see the 'Methods' section