Coupled European and Greenland last glacial dust activity driven by North Atlantic climate

Újvári, Gábor; Stevens, Thomas; Molnár, Mihály; Demény, Attila; Lambert, Fabrice; Varga, György; Timothy Jull, A. J.; Páll-Gergely, Barna; Buylaert, Jan Pieter; Kovács, János

Published in:
Proceedings of the National Academy of Sciences of the United States of America

Link to article, DOI:
10.1073/pnas.1712651114

Publication date:
2017

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):
Újvári, G., Stevens, T., Molnár, M., Demény, A., Lambert, F., Varga, G., Timothy Jull, A. J., Páll-Gergely, B., Buylaert, J. P., & Kovács, J. (2017). Coupled European and Greenland last glacial dust activity driven by North Atlantic climate. Proceedings of the National Academy of Sciences of the United States of America, 114(50), E10632-E10638. https://doi.org/10.1073/pnas.1712651114

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.
Coupled European and Greenland last glacial dust activity driven by North Atlantic climate

Gábor Újvári,a,b,1 Thomas Stevens,c Mihály Molnár,d Attilla Demény,e Fabrice Lambert,f György Varga,f A. J. Timothy Jull,g,h Barna Pál-Gergely,i Jan-Pieter Buylaert,b,i and János Kovácsa,b

1Institute for Geological and Geophysical Research, Research Centre for Astronomy and Earth Sciences, Hungarian Academy of Sciences, H-1112 Budapest, Hungary; 2Center for Nuclear Technologies, Technical University of Denmark, 4000 Roskilde, Denmark; 3Department of Earth Sciences, Uppsala University, 75236 Uppsala, Sweden; 4Isotope Climatology and Environmental Research Centre, Institute for Nuclear Research, Hungarian Academy of Sciences, H-4026 Debrecen, Hungary; 5Geography Institute, Pontificia Universidad Católica de Chile, 7810000 Santiago, Chile; Geographical Institute, Research Centre for Astronomy and Earth Sciences, Hungarian Academy of Sciences, H-1112 Budapest, Hungary; 6Department of Geosciences, University of Arizona, Tucson, AZ 85721; 7University of Arizona Accelerator Mass Spectrometry Facility, University of Arizona, Tucson, AZ 85721; 8Plant Protection Institute, Centre for Agricultural Research, Hungarian Academy of Sciences, H-1022 Budapest, Hungary; 9Nordic Laboratory for Luminescence Dating, Department of Geoscience, University of Aarhus, 4000 Roskilde, Denmark; 10Department of Geology and Meteorology, University of Pécs, H-7624 Pécs, Hungary; and 11Environmental Analytical and Geoanalytical Research Group, Széntágothai Research Centre, H-7624 Pécs, Hungary

Edited by Mark H. Thiemens, University of California, San Diego, La Jolla, CA, and approved October 26, 2017 (received for review July 15, 2017)

Centennial-scale mineral dust peaks in last glacial Greenland ice core is the timing of lowest Greenland temperatures, yet little is known of equivalent changes in dust-emitting regions, limiting our understanding of dust–climate interaction. Here, we present the most detailed and precise age model for European loess dust deposits to date, based on 125 accelerator mass spectrometry 14C ages from Dunaszekcsó, Hungary. The record shows that variations in glacial dust deposition variability on centennial–millennial timescales in east central Europe and Greenland were synchronous within uncertainty. We suggest that precipitation and atmospheric circulation changes were likely the major influences on European glacial dust activity and propose that European dust emissions were modulated by dominant phases of the North Atlantic Oscillation, which had a major influence on vegetation and local climate of European dust source regions.

dust | Dansgaard–Oeschger events | European loess | radiocarbon dating | mollusk shell

Atmospheric mineral dust (hereafter called dust) plays a major but poorly understood role in the climate system, both responding to and driving climate change (1). Dust concentrations in the Greenland ice cores show a close relationship to Greenland temperature at millennial to centennial timescales, although the precise causes and effects of this relationship remain unclear (2). At millennial to centennial timescale, last glacial climate variability over Greenland, and likely over the North Atlantic generally, is dominated by the Dansgaard–Oeschger (D–O) cycles (3, 4). In Greenland, each D–O cycle over the last 120 ka is characterized by (i) gradual cooling over a century to many millennia; (ii) subsequent, more rapid decline into peak stadial conditions; and (iii) very abrupt return to interstadial conditions, varying in amplitude from 5 K to 16.5 K (5, 6). Several mechanisms behind these abrupt climatic changes have been proposed, including ocean circulation (7), sea ice cover–ice sheet (8, 9), and wind pattern changes (10). The effects of D–O events extended across much of the Northern Hemisphere (11) and were identified in precisely dated terrestrial records such as speleothems in Europe and Asia (12–15).

However, changes in climate and dustiness in potential dust-emitting source regions are less well understood at millennial scales (16). Furthermore, recent isotopic and clay mineral evidence from Northern Hemisphere loess deposits reopened the debate about the origins of Greenland dust, as both East Asian and European dust sources are compatible with current Greenland dust data (17). Modeling studies highlight that changes in precipitation patterns and vegetation cover over these potential source areas profoundly affected the dust cycle in response to D–O–Heinrich events (18, 19), but well-dated, high-resolution dust records covering parts of or the whole last glacial period are scarce. Some studies on extensive windblown terrestrial dust deposits of loess that cover large areas of land surface have suggested imprints of the D–O events exist in loess grain size records in Europe (20, 21) and Asia (22). However, the chronologies of these archives are often poor or not fully independent. Furthermore, the vast majority of fully independently dated loess sequences are dated using luminescence methods (23, 24). Although these methods generally provide accurate age estimates for Late Quaternary deposits, they do not have sufficient precision to investigate the timing and underlying mechanisms of millennial–centennial-scale environmental changes, including dust accumulation variability on the continents (25, 26).

Thus, despite being globally widespread and closely tied to or acting as major global dust source regions, loess records have so far remained a largely untapped source of information on dustiness at millennial timescales and below. As other dust records tend to be geographically isolated and restricted in area, this has been a major impediment to understanding cause and effect in climate–dustiness feedbacks. Recent advances in high-precision accelerator mass spectrometry (AMS) radiocarbon dating of small gastropods in loess (26–28) provide a new opportunity to gain unprecedented insights into the temporal variations of the terrestrial dust cycle at millennial to centennial scale from loess.

Significance

Atmospheric dust is a major component of climate change. However, the relationship between glacial continental dust activity and abrupt centennial–millennial-scale climate changes of the North Atlantic is poorly known. Recent advances in high-precision radiocarbon dating of small gastropods in continental loess deposits provide an opportunity to gain unprecedented insights into dust variations and its major drivers at centennial–millennial scales from a near-source dust archive. Here, we show that Late Quaternary North Atlantic temperature and dustiness in Greenland and Europe were largely synchronous and suggest that this coupling was driven via precipitation changes and large-scale atmospheric circulation.

Author contributions: G.Ú. designed research; G.Ú. performed research; M.M., A.D., A.J.T.J., and J.K. contributed new reagents/analytic tools; G.Ú., M.M., A.D., G.V., A.J.T.J., B.P.-G., J.P.B., and J.K. analyzed data; and G.Ú., T.S., and F.L. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

This open access article is distributed under Creative Commons Attribution-NonCommercial-NoDerivatives License 4.0 (CC BY-NC-ND).

1To whom correspondence should be addressed. Email: ujvari.gabor@csfk.mta.hu.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1712651114/-/DCSupplemental.
results of a recent radiocarbon dating study of earthworm granules from soils in a loess site in Germany points to a link between Greenland climate and soil formation in western European loess (29). Although this highlights the potential of radiocarbon dating of loess, it is not clear whether the inferred precipitation changes in that study are accompanied by shifts in dust accumulation, crucial to understanding past dustiness. Here we present a high-resolution loess grain size and dust accumulation record from southern Hungary covering the period of 36 to 24 ka before 2000 (b2k) with a fully independent radiocarbon chronology based on a uniquely detailed age dataset consisting of 125 AMS $^{14}$C dates and an associated Bayesian age–depth model. Our time resolution allows investigating the timing of abrupt European climate shifts compared with the North Greenland Ice Core Project (NGRIP) Ca$^{2+}$ record in Greenland (S). We also measured species-specific mollusc shell carbon and oxygen isotopic data as potential indicators of coupled precipitation–vegetation changes, which may be a major driver of continental dust production and deposition variations in east central Europe at centennial–millennial timescales.

**Results**

**Site and Chronology.** The Dunaszekcső loess–paleosol sequence is situated in southern Hungary (Dsz, Fig. 1). This part of the Carpathian Basin is an area of low relief between the main mountain ranges of central Europe and is under Atlantic, Mediterranean, and continental climatic influence. This is expressed in the amount of annual rainfall (375 mm y$^{-1}$, with extremes of 270 mm y$^{-1}$ to 882 mm y$^{-1}$) and mean air temperatures during winter ($\sim$0.3 °C, January) and summer ($\sim$22.2 °C, July) as measured at a nearby meteorological station for the period 1998–2013. Back-trajectory analyses revealed five major sources of present-day precipitation at K-puszta, a site on the Great Hungarian Plain: Mediterranean region (57.0%), local moisture (14.8%), Atlantic region (14.2%), north Europe (7.4%), and east Europe (6.6%) (30).

The exposed section of the Dsz record (ca. 17 m) is the upper part of a ~70-m-thick Quaternary aeolian sediment sequence and covers the last glacial–interglacial cycle (26). Sediment samples were collected at 5-cm resolution between 250 and 1,045 cm in the profile for dating and proxy analyses (see Methods). As shown by calibrated AMS $^{14}$C ages of numerous charcoal fragments and mollusk shells (Datasets S1 and S2), the sampled profile covers a period of loess sedimentation between 36 cal y BP and 23.4 cal y BP. Radiocarbon ages of charcoals and small mollusks are consistent despite the very different origin and genesis of these materials (26, 28), and demonstrate that the age dataset is reliable and robust. The Bayesian age–depth model of the composite Dsz record is calculated at 1-cm intervals and has a minimum 95% age uncertainty of 188.7 y at 486 cm (25.796 cal y BP) and a maximum of 1,328.7 y at 1,045 cm (35.988 cal y BP) (Figs. S1–S3). In the older section, charcoal $^{14}$C ages become less precise as they approach the practical limit of radiocarbon dating.

**Sedimentation—Mass Accumulation Rates and Grain Size.** Sedimentation rates have changed significantly over time, as indicated by relatively low values (mean: $\sim$0.4) of the posterior distribution for memory (or autocorrelation) after Bayesian age modeling using Bacon software (31) (Figs. S2 and S3). We therefore infer that the environmental conditions or internal dynamics of the aeolian system that influence loess accumulation were highly variable at the study site. Bulk mass accumulation rates (BMARs) and dust mass accumulation rates (DMARs; $<10$ μm dust fraction) range between 380 g m$^{-2}$ y$^{-1}$ and 2,885 g m$^{-2}$ y$^{-1}$ (1-cm-resolution model) and 69 g m$^{-2}$ y$^{-2}$ and 629 g m$^{-2}$ y$^{-2}$ (5-cm-resolution model), respectively, showing an almost 10-fold increase between the minimum at 32.6 ka b2k and the maximum at 26.1 ka b2k (1-cm-resolution model, Dataset S2). BMAR and DMAR display a longer-term increasing trend, following insolation at 45°N, and exhibit centennial- to millennial-scale variability corresponding, in general, to the stadial–interstadial pattern recorded in Greenland (Fig. 2). Significant decreases and increases in BMAR occur within short periods of time, generally coeval with the NGRIP dust record (Figs. 2 and 3).

Median particle diameters of bulk loess ($d_{50_{bulk}}$) and quartz in loess ($d_{50_{quartz}}$) vary between 18.5 μm to 55.7 μm and 22.4 μm to 64.3 μm, respectively, and both exhibit centennial-scale variability. While quartz grain size does not show an insolation-driven trend, bulk grain size displays a general coarsening after $\sim$30 ka b2k to 31 ka b2k. A striking feature of the grain size records is that they are not characterized by consistent coarsening–fining trends that match Greenland Stadial–Greenland Interstadial (G5/G1) patterns, and the two records often show opposite trends to the Dsz BMAR and DMAR, and the NGRIP Ca$^{2+}$ records (e.g., G1-5.1 and G1-4, Fig. 2).

**Discussion**

Comparison of dust accumulation in Greenland and Hungary reveals some striking patterns on orbital, millennial, and centennial timescales, which we suggest demonstrates close and potentially causative links between North Atlantic climate and European dust activity. Over orbital timescales, there is a pattern of increasing BMAR in the Dsz record after 31 ka b2k (Fig. 2), which corresponds to decreasing spring–summer insolation at 45°N latitude, culminating in the highest dust inputs during the early Last Glacial Maximum (LGM). This pattern is broadly consistent with the Greenland Ca$^{2+}$ dust record, although the latter shows less of a marked increase at the LGM and this increase seems to last longer (S). The LGM was an especially cold and dry period in the North Atlantic and Europe (34, 35), accompanied by changes in European vegetation cover (36) and different atmospheric circulation patterns driven by profound changes in insolation (32), topography (ice sheets, ref. 37) and trace gas compositions (34). The orbital timescale changes in the dust cycle, as recorded in the Dsz sequence, were likely driven by insolation changes and longer term cooling–drying trends that resulted in surface conditions appropriate for more intense dust emissions in Europe during the LGM.

Beyond this first-order insolation control, centennial–millennial timescale variability in Dsz BMAR and DMAR also became more pronounced from 31 ka b2k onward. Some traces of these higher frequency oscillations can be observed before 31 ka, although the
Fig. 2. Comparison of insolation and paleoclimate records for the 37- to 22-ka interval. (A) Insolation for June 21 at 45°N (32), and (B) integrated insolation for the spring season at 45°N (33). (C) Median grain size of quartz grains (d50quartz) in the Dsz loess record (5-cm resolution). (D) Median grain size of bulk loess (d50bulk) in the Dsz loess record (5-cm resolution). (E) DMARs of loess at Dsz calculated from the lower (5 cm) resolution age model. (F) BMARs at Dsz calculated from the higher (1 cm) resolution Bayesian age–depth model. (G) Sieben Hängste (7H) composite stalagmite δ18O record (Western Alps) (15). (H) and (I) NGRIP dust and ice/water δ18O records (5). Yellow bars denote GI periods as given by Rasmussen et al. (5), while gray bars indicate phases of reduced Ca\(^{2+}\) (less dust) within GS periods.

precise timing becomes more uncertain because of the lower number of radiocarbon ages from this part of the sequence and the fact that the age model is less precise at this point. In general, the high-precision \(^{14}C\) chronology and BMAR record at Dunaszke
cso allows unprecedented insights into the timing of abrupt European dust cycles changes and a detailed comparison with Greenland climate and dust. A striking feature is that Dsz dust accumulation minima generally match GI phases (GI-5.1, GI-4, and GI-3; Fig. 3) in the NGRIP δ18O/Ca\(^{2+}\) records (5) within dating uncertainties, and also with shorter periods of low Ca\(^{2+}\) during the LGM (gray bars, Figs. 2 and 3). While the timing of Dsz BMAR decrease and subsequent increase nicely matches the onset and end of GI-5.1 in the NGRIP record, the onset of GI-4 is missing in the Dsz record. This is because BMAR stays relatively low (at interstadial levels) after a doubling from ~600 g m\(^{-2}\) y\(^{-1}\) to ~1,000 g m\(^{-2}\) y\(^{-1}\) at ~30.6 ka b2k to 30.5 ka b2k. The DMAR minimum around 29 ka b2k as well as the subsequent abrupt increase in BMAR and DMAR are within dating uncertainty of their GI-4 counterparts in the Greenland record (Figs. 2 and 3). Similarly, a DMAR and BMAR minimum is synchronous with the Greenland GI-3 within dating uncertainty. Significant increases in dust sedimentation occur during GS phases, with BMAR peaks reaching and exceeding 2,500 g m\(^{-2}\) y\(^{-2}\) between ~25.8 ka b2k and 26 ka b2k. This maximum matches the timing of peak dust accumulation over central Greenland (Figs. 2 F and H and 3 A), although the highest dust accumulations in Dsz are split into two distinct peaks. By contrast, grain size variations (d50bulk and d50quartz) do not necessarily coincide with BMAR, DMAR, and the GI/GS variability, and tend to show coarsening during GI periods (e.g., GI-5.1 and GI-4; Fig. 2 C and D), when the opposite would be expected. Mismatches between grain size and dust accumulation rates (AR) were also found at Crvenka in Serbia (23) and at sites on the Chinese Loess Plateau, using luminescence dating (38). This result contrasts with previous observations at Nussloch, Germany, where layers of coarse (fine) loess grain size have been suggested to correspond to GS (GI) phases in Greenland (20, 21). This may reflect the differences in aeolian sedimentation between a more humid, western European site, where grain size...
is largely controlled by soil formation, and those in drier east central–SE Europe where grain size can be directly linked with aeolian transport, trapping, and deposition. The general disagreement between the BMAR, DMAR, and δ50bulk,quartz proxies is likely due to the fact that grain size is a much more complex parameter on short timescales. Grain size distributions (GSDs) are affected by multiple, often stochastic processes, and the clay- to sand-sized particles making up loess are mobilized, transported, and deposited in many different ways (39). Conversely, BMAR only reflects the amount of particles deposited per unit time and area, which is a function of atmospheric loading, local trapping, and preservation conditions. In this study, we will therefore refrain from using grain size to identify millennial- to centennial-scale events in loess deposits.

The match between the Dunaszekcső and Greenland dust records can be further analyzed within a wider European context. The onset of GI-5.1 in Greenland is synchronous with the reduction in dust accumulation in southern Hungary within the age scale uncertainties, and there is an excellent match between GI-5.1 and the dust minimum at ~30.7 ka b2k (Fig. 3). While BMAR stays relatively low (still in the range of 600 g m⁻² y⁻¹ to 1,000 g m⁻² y⁻¹) after GI-5.1, a significant increase in dust input starts at 28.8 ka b2k, coincident with a drop in δ¹⁸O of the 7H Alpine stalagmite composite record, and the end of GI-4 within dating uncertainty (Fig. 3; note that 2σ error at 28.8 ka is 574 y for Dunaszekcső). The onset of GI-3 is coeval in Greenland and the Dsz record, and the duration of the dust flux decrease in the Dsz record is longer than that of GI-3 (240 y) (5). At the same time, the 7H record exhibits the shortest D-O event at ~27.7 ka b2k. Further smaller decreases in dust accumulation at Dsz occurred in line with similar events in Greenland, which can also be found in the 7H record within dating uncertainty (Fig. 3, gray bars). Unfortunately, the combined uncertainty of the Dsz age model together with the IntCal13 calibration uncertainties (IntCal13 is the latest calibration curve published by the IntCal group of the international radiocarbon community, ref. 40) preclude the identification of any lead or lag between the various records. Nevertheless, the observation that there is a lot of shared variability between NGRIP and Dsz over 32 ka b2k to 24 ka b2k suggests a good general agreement between the ice core and IntCal13 chronologies and may potentially indicate a possible European dust contribution to Greenland ice cores, an issue that has not yet been satisfactorily resolved from provenance studies and has important implications for dust transport pathways (17).

Fig. 3. Dust accumulation variations in Greenland and east central Europe and precipitation patterns in the Western Alps for the 32- to 24-ka period. (Lower) Records shown are (A) high-resolution BMAR in southern Hungary (Dunaszekcső record), (B) the 7H stalagmite δ¹⁸O record (15), and (C) NGRIP dust (Ca²⁺) (5). (Upper) The 2-sigma age model (or counting) uncertainties of the displayed records. Yellow and gray bars indicate decrease in dust accumulation over Greenland. Dust minima identified as interstadials in the NGRIP record are marked by yellow bars (GI-5.1, GI-4, and GI-3; ref. 5).
The match between the Dsz BMAR and 7H δ18O records indicates that large-scale atmospheric reorganizations are the likely cause of dust cycle changes in east central Europe. We here propose that the predominant modes of the North Atlantic Oscillation (NAO) during the LGM and resultant changes in storm tracks may explain these patterns. The 7H speleothem δ54Si dataset reflects precipitation patterns and reveals intense southern moisture advection from the Mediterranean toward the Alps during the LGM, related to Rossby wave breaking of the jet stream over western Europe (15). A related southward shift in the position of the North Atlantic storm track was also found in recent modeling studies of LGM atmospheric circulation (37, 41, 42), explained by a tendency toward more cyclonic wave breaking (CBW) and less anticyclonic wave breaking (AWB) events in the LGM runs (41, 42). Importantly, under current climate, teleconnections such as the NAO (characterized by a meridional displacement of the upper-tropospheric Atlantic jet; ref. 43) have a dynamic link to Rossby wave breaking events (44). The negative (positive) phase of the present-day NAO relates to a more southward (northward) position of the eddy-driven Atlantic jet, and exhibits more CBW (AWB) than the long-term average (42). The effect of this dynamic link on climate is stark, as it affects seasonal and, in particular, wintertime temperature and precipitation patterns in Europe. For example, during the prolonged Maunder Minimum cold period from 1640 to 1715, associated with an NAO− phase, winters were characterized by more-frequent blocking situations connected with cold air outbreaks toward central and eastern Europe (45). Springs were cold and characterized by a southward shift of the midlatitude storm tracks, and summers in central Europe were wetter and slightly cooler than they are today. Simulations performed using a coupled atmosphere–ocean model for the Maunder Minimum revealed higher cyclone density in central–east central Europe for both the winter and spring periods (46), which were also found to be the major dust emission periods in east central Europe under cold climate states during the last glaciation (February–June; ref. 18). In contrast to the Maunder Minimum cold period, a persistent NAO+ mode was reconstructed for the Medieval Warm Period from ~800 to 1300 (47). Modeling studies suggest that the NAO during the LGM differed from the modern one, as it was characterized by four centers of action (48), and a latitudinal fluctuation of the eddy-driven jet (42). However, some simulations demonstrate that latitudinal wobbling remains the primary type of North Atlantic jet variability and also dominates jet pulsing during glacial times (49). As such, we suggest that the dust minima in the Dsz BMAR and DMAR records during Greenland GI events could be explained by a prolonged NAO+ mode with a more northerly jet position (NW trajectory in the 7H composite record, Fig. 3). Conversely, dust maxima during GS events would be linked to a persistent NAO− phase. Spring δ18O− phase, eastern European summers would have been drier, benefitting C4 vegetation. NAO− and NAO+ phase dominant periods co-incide with Greenland GS and GI, respectively.

Methods

Site and Sampling. Sampling for grain size analyses was performed at 5 cm resolution on two overlapping profiles (Fig. S1) of a carefully cleaned loess-paleosol sequence at Dunaszentkőz (southern Hungary, 46°05′25″N, 18°45′45″E, 135 m above sea level), freshly exposed by a landslide in 2008. AMS radiocarbon dating was conducted on samples collected at 5-cm-depth resolution between the depths of 485 and 850 cm and at 15- to 30-cm-depth resolution for the rest of the sequence. For 14C-dating, loess cuboids with dimensions of 15 × 5 × 5 cm were prepared and combusted in one step at 1,000 °C (ABA-OSC100g). While, during subsequent runs, they were subjected to stepped combustion in a pure O2 gas atmosphere, first at 400 °C and then at 800 °C (ABA-TSC100 and ABA-TSC200) (28). Two samples with low carbon yields were measured via a gas ion source attached to the AMS.

AMS Radiocarbon Dating. Before the 14C measurements, charcoal fragments were treated using the standard acid-base–acid (ABA) method. After the final acid wash, the samples were washed again with distilled water to neutral pH and freeze-dried overnight. First, dried charcoal fragments were combusted in an on-line combustion system using CuO in one step at 1,000 °C (ABA-OSC100g), while, during subsequent runs, they were subjected to stepped combustion in a pure O2 gas atmosphere, first at 400 °C and then at 800 °C (ABA-TSC100 and ABA-TSC200) (28). Two samples with low carbon yields were measured via a gas ion source attached to the AMS.

Mollusk shells were ultrasonically washed and etched using weak acid (2% HCl) to remove surface contaminations (20 to 30% of the original mass removed). Subsequently, mollusk shells were dried and put in vacuum-tight two-finger reaction ampoules (~100 cm3 inner volume) and dissolved using phosphoric acid. CO2 was produced by acid hydrolysis of shells at 75 °C, further purified cryogenically, and then graphitized (50).

All of the 14C measurements were undertaken using a compact radiocarbon AMS system (MCADAS) at the Hertelendi Laboratory of Environmental Studies, Institute for Nuclear Research, Debrecen (Hungary). Conventional radiocarbon ages were converted to calendar ages using OxCal online (version 4.2; ref. 51) and the IntCal13 calibration curve (46).

Composite Profile. The composite profile was obtained using the AMS 14C ages and bulk median diameter (d50) values measured in profiles 1 and 2 (Fig. S1). The validity of this approach was subsequently checked by Monte Carlo simulation with the “Intra-site correlation age modelling” code (52).

Age–Depth Modeling. Bayesian age–depth modeling was performed using the Bacon code (31), based on 125 radiocarbon data points (Dataset S1). Inverse AR (sedimentation times, years per centimeter) were estimated from 35.6 million Markov Chain Monte Carlo (MCMC) iterations, and these rates form the age-depth model. Inverse AR were first constrained by default prior information: accumulation (acc.) = 1.5 and acc.mean = 20 for the gamma distribution, and memory (mem.mean) = 0.7 and mem.size = 4 for the beta distribution describing the memory effects (or autocorrelation) of inverse AR. All input age data were provided as 14C y BP, and Bacon used the IntCal13 calibration curve to convert conventional radiocarbon ages to calendar ages. Age modeling was run to achieve a 5-cm final resolution. In a second attempt, as part of a sensitivity test, the parameters were set as acc.shape = 2 and acc.mean = 20 for the gamma distribution and mem.mean = 0.4 and mem.size = 17 based on the posterior gamma and beta distributions from the first modeling attempt (Fig. S2). Posterior gamma and beta distributions better fitted the priors for the second model (Fig. S3), and the mean of inverse AR after modeling was found to be ~25 y cm−1, matching very well the regional average (26.55 y cm−1) of east central European loess records (Table S1). Although only insignificant differences were found between these age models (Figs. S4 and S5), the second age–depth model was used to interpret proxy data in this study (Dataset S2). Finally, the age modeling was run again using the Bacon code with a 1-cm-mesh model (Dataset S2). To make the age scale comparable to the ice core timescale (GICC05), 50 y were added. All ages are therefore presented in figures in ice core years b2k (before year AD 2000).

E10636 | www.pnas.org/cgi/doi/10.1073/pnas.1712651114

Újvári et al.
Mass Accumulation Rate Calculations. AR were calculated using the age–depth model as AR (meters per year) = d – d0, where d0 and d are carbonate-weighted mean ages in the profile. BMRAs have been computed using the equation BMR (grams per square meter per year) = AR × f × f, where f is the dry bulk density (grams per cubic meter), and f is the sediment fraction that is aeolian in origin. Since loess is

This study was funded by Hungarian National Research, Development and Innovation Office Grant OTKA PD-108639 (to G.U.). Additional financial support provided by Boljai János Research Scholarship of the Hungarian Academy of Sciences Project BO/00326/15 (to G.U.) is acknowledged. The project has been also supported by the European Union, cofinanced by European Social Fund Grant EFOP-3.6.1-16-2016-00004 (to J.K.).

Grain Size Analyses. Before laser diffraction measurements, 3 g of loess samples were pretreated with 10 mL of 20% H2O2 and 10 mL of 10% HCl to remove organic matter and carbonates. Subsequently, 10 mL NaPO4 was added to the samples, which were finally ultrasonicated for ~1 min. These are chemically fully dispersed samples and the presented bulk loess grain size data are based on fully dispersed PSDs.

To obtain pure quartz separates, air-dried bulk sediment samples (2 g each) were first treated with 10 mL of 10% HCl for 24 h to remove organic matter. Subsequently, 50 mL of 6 M HCl was added, and the solution was boiled at 90 °C for 1 h to remove carbonates and iron oxides. Quartz was isolated by the sodium pyrosulfate fusion–hydrofluoric acid method. This procedure resulted in pure quartz separates without affecting the grain size, grain shape, and surface textures of the quartz crystals, as has been confirmed by SEM imaging.

Grain size of bulk loess samples and quartz separates was analyzed using a Malvern Instruments Masterizer 3000 laser diffractometer with a Hydro LV wet dispersion unit having a measurement range of 0.1 μm to 2,100 μm divided into 100 size bins. Two light sources were utilized, a red He-Ne laser at a wavelength of 0.633 mm and a blue LED at 0.470 mm. Diffraction light intensity was measured by 50 sensors over a wide range of angles. Constants of 1.33 for the refractive index of water, 1.544 for the refractive index of solid phases (valid for quartz, and most clay minerals and feldspars), and an absorption index of 0.1 were applied for both bulk loess and quartz. Bulk grain size analyses reported in this paper are the average of seven successive laser diffraction runs (total of 70,000 snaps), while those of quartz are the mean of three repeat measurements. The recorded data were processed using the Malvern’s Masterizer 3000 software (version 3.10), which transformed the scattered light data to particle size information based on the Mie Scattering Theory. Median grain size of bulk loess samples (d50) and quartz separates (d50 quartz) was calculated from the Masterizer 3000 software outputs.

ACKNOWLEDGMENTS. This study was funded by Hungarian National Research, Development and Innovation Office Grant OTKA PD-108639 (to G.U.). Additional financial support provided by Boljai János Research Scholarship of the Hungarian Academy of Sciences Project BO/00326/15 (to G.U.) is acknowledged. The project has been also supported by the European Union, cofinanced by European Social Fund Grant EFOP-3.6.1-16-2016-00004 (to J.K.).
45. Luterbacher J, et al. (2001) The late Maunder Minimum (1675–1715)—A key period for studying decadal scale climatic change in Europe. Clim Change 49:441–462.
46. Raible CC, Yoshimori M, Stocker TF, Casty C (2007) Extreme midlatitude cyclones and their implications for precipitation and wind speed extremes in simulations of the Maunder Minimum versus present day conditions. Clim Dyn 28:409–423.
47. Trouet V, et al. (2009) Persistent positive North Atlantic Oscillation mode dominated the Medieval Climate Anomaly. Science 324:78–80.
48. Justino F, Peliter WR (2005) The glacial North Atlantic Oscillation. Geophys Res Lett 32:L21803.
49. Mertz N, Raible CC, Woolings T (2015) North Atlantic eddy-driven jet in interglacial and glacial winter climates. J Clim 28:3977–3997.
50. Molnár M, et al. (2013) Status report of the new AMS $^{14}$C sample preparation lab of the Hertelendy Laboratory of Environmental Studies (Debrecen, Hungary). Radiocarbon 55:665–676.
51. Bronk Ramsey C (2009) Bayesian analysis of radiocarbon dates. Radiocarbon 51:337–360.
52. Fohlmeister J (2012) A statistical approach to construct composite climate records of dated archives. Quat Geochronol 14:48–56.
53. Újvári G, Kovács J, Varga Gy, Raucsik B, Markovic SB (2010) Dust flux estimates for the Maunder Minimum versus present day conditions. Earth Planet Sci Lett 28:409–423.
54. Justino F, Peltier WR (2005) The glacial North Atlantic Oscillation. Geophys Res Lett 32:L21803.
55. Luterbacher J, et al. (2001) The late Maunder Minimum (1675–1715)—A key period for studying decadal scale climatic change in Europe. Clim Change 49:441–462.
56. Újvári G, Kovács J, Varga Gy, Raucsik B, Markovic SB (2010) Dust flux estimates for the Maunder Minimum versus present day conditions. Earth Planet Sci Lett 28:409–423.
57. Varga Gy, Raucsik B, Markovic SB (2010) Dust flux estimates for the Maunder Minimum versus present day conditions. Earth Planet Sci Lett 28:409–423.
58. Bronk Ramsey C (2009) Bayesian analysis of radiocarbon dates. Radiocarbon 51:337–360.
59. Luterbacher J, et al. (2001) The late Maunder Minimum (1675–1715)—A key period for studying decadal scale climatic change in Europe. Clim Change 49:441–462.
60. Újvári G, Kovács J, Varga Gy, Raucsik B, Markovic SB (2010) Dust flux estimates for the Maunder Minimum versus present day conditions. Earth Planet Sci Lett 28:409–423.
61. Antoine P, et al. (2009) High-resolution record of the last climatic cycle in the eastern part of the Polish Carpathians. Folia Malaarol 13:49–94.
62. Hatté C, et al. (2013) Excursions to C4 vegetation recorded in the Upper Pleistocene loess of Surduk (Northern Serbia): An organic isotope geochemistry study. Clim Past 9:1001–1014.
63. Schmidt ED, Machesett B, Markovic SB, Tsukamoto S, Frechen M (2010) Luminescence chronology of the upper part of the Stari Slankamen loess sequence (Vojvodina, Serbia). Quat Geochronol 5:137–142.
64. Kehrwald N, McCoy WD, Thibeault J, Burns SJ, Oches EA (2010) Palaeoclimatic implications of the spatial patterns of modern and LGM European land-sand shell $^{14}$C. Quat Res 74:166–176.
65. Landis GP (1983) Harding Iceland Spar: A new $^{18}$O/$^{16}$O carbonate standard for hydrothermal minerals. Isot Geosci 1:91–94.
66. Welter-Schultes FW (2012) European Non-Marine Molluscs, a Guide for Species Identification (Planetary Poster Ed, Göttingen, Germany).
67. Sümegi P, Krollopp J (2002) Quaternary palaeoecological analysis for modeling of the Upper Weichselian palaeoenvironmental changes in the Carpathian Basin. Quat Int 91:53–63.
68. Prockov M, Drivotová M, Juríčková L, Kuznik-Kowalska E (2013) Field and laboratory studies on the life-cycle, growth and feeding preference in the hairy snail Trochulus hispidus (L., 1758) (Gastropoda: Pulmonata: Hygromiidae). Biology 68:131–141.
69. Sulikowska-Drozd A (2005) Distribution and habitat preferences of clausiliids (Gastropoda: Pulmonata: Clausiliidae) in the eastern part of the Polish Carpathians. Folia Malacol 30:139–94.
70. Scott L (2002) The influence of diet on the $^{13}$C of shell carbon in the pulmonate snail Helix aspersa. Earth Planet Sci Lett 195:249–259.
71. Metref S, Rousseau D, Bântel B, Labonne M, Vianey-Liaud M (2003) Study of the diet effect on $^{13}$C of shell carbonate of the land snail Helix aspersa in experimental conditions. Earth Planet Sci Lett 211:381–393.
72. Balakrishnan M, Yapp C (2004) Ox in balance models for the oxygen and carbon isotope compositions of land snails. Earth Planet Sci Lett 211:381–393.
73. Lécolle P (1985) The oxygen isotope composition of landsnail shells as a climatic indicator. Palaeogeogr Palaeclim Palaeoecol 23:93–101.
74. Zanchetta G, Leone G, Fallick A, Bonadonna F (2005) Oxygen isotope composition of living land snail shells: Data from Italy. Palaeogeogr Palaeclim Palaeoecol 223:20–33.
75. Goodfriend G, Magaritz M (1987) Carbon and oxygen isotope composition of land snail shells. Geochim Cosmochim Acta 68:2007–2024.
76. Lécoult P (1985) The oxygen isotopes composition of landsnail shells as a climatic indicator. Appl hydrogeology and paleoclimatology. Chem Geol 58:157–181.
77. Zanchetta G, Leone G, Fallick A, Bonadonna F (2005) Oxygen isotope composition of living land snail shells: Data from Italy. Palaeogeogr Palaeclim Palaeoecol 223:20–33.
78. Goodfriend G, Magaritz M (1987) Carbon and oxygen isotope composition of land snail shells. Earth Planet Sci Lett 86:377–388.
79. Balakrishnan M, Yapp C, Theler J, Carter BJ, Wyckoff DG (2005) Environmental significance of C14 and O18 ratios of modern land snail shells from the southern great plains of North America. Quat Res 63:15–30.