Synchronizing ¹⁰Be in two varved lake sediment records to IntCal13 ¹⁴C during three grand solar minima

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Abstract. Time-scale uncertainties between paleoclimate reconstructions often inhibit studying the exact timing, spatial expression and driving mechanisms of climate variations. Detecting and aligning the globally common cosmogenic radionuclide production signal via a curve fitting method provides a tool for the quasi-continuous synchronization of paleoclimate archives. In this study, we apply this approach to synchronize ¹⁰Be records from varved sediments of Lakes Tiefer See and Czechowskie covering the Maunder-, Homeric- and 5500 a BP grand solar minima with ¹⁴C production rates inferred from the IntCal13 calibration curve. Our analyses indicate best fits with ¹⁴C production rates when the ¹⁰Be records from Lake Tiefer See were shifted for 8 (-12/+4) (Maunder Minimum), 31 (-16/+12) (Homeric Minimum) and 86 (-22/+18) years (5500 a BP grand solar minimum) towards the past. The best fit between the Lake Czechowskie ¹⁰Be record for the 5500 a BP grand solar minimum and ¹⁴C production was obtained when the ¹⁰Be time-series was shifted 29 (-8/+7) years towards present. No significant fits were detected between the Lake Czechowskie ¹⁰Be records for the Maunder- and Homeric Minima and ¹⁴C production, likely due intensified in-lake sediment resuspension since about 2800 a BP, transporting 'old' ¹⁰Be to the coring location. Our results provide a proof of concept for facilitating ¹⁰Be in varved lake sediments as novel synchronization tool required for investigating leads and lags of proxy responses to climate variability. However, they also point to some limitations of ¹⁰Be in these archives mainly connected to in-lake sediment resuspension processes.

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1. Introduction

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Paleoclimate archives provide unique insights into the dynamics of the climate system under various forcing conditions (Adolphi et al., 2014; Brauer et al., 2008; Neugebauer et al., 2016). Particularly the timing and spatial expression of climate variations can provide valuable information about the underlying driving mechanisms (Czymzik et al., 2016b, 2016c; Lane et al., 2013; Rach et al., 2014). However, time-scale uncertainties between different paleoclimate records often inhibit the investigation of such climate variations. Climate independent synchronization tools offer the possibility for synchronizing individual paleoclimate archives and, thereby, robust studies of leads and lags in the climate system.

In addition to volcanic tephra layers (Lane et al., 2013), atmospheric trace gases (Pedro et al., 2011) and paleomagnetism (Stanton et al., 2010), cosmogenic radionuclides like ¹⁰Be and ¹⁴C provide such a synchronization tool (Adolphi et al., 2017; Adolphi and Muscheler, 2016). The isotopes are produced mainly in the stratosphere through cascades of nuclear reactions triggered by incident high energy galactic cosmic rays (Lal and Peters, 1967). The flux of these galactic cosmic rays into the atmosphere is, in turn, modulated on up to multi-centennial-scales mainly by solar activity changes (Stuiver and Braziunas, 1989). On >500-year intervals further cosmogenic radionuclide production changes induced by the varying geomagnetic field become increasingly important (Lal and Peters, 1967; Snowball and Muscheler, 2007; Simon et al., 2016). Detecting and aligning the externally forced cosmogenic radionuclide production signal via a curve fitting method enables the quasi-continuous synchronization of natural environmental archives (Adolphi and Muscheler, 2016; Muscheler et al., 2014).

One challenge with this approach is the unequivocal detection of the cosmogenic radionuclide production signal because of transport and deposition processes. Subsequent to production, ¹⁴C oxidizes to ¹⁴CO₂ and enters the global carbon cycle. Varying exchange rates between Earth's carbon reservoirs add non-production variability to the atmospheric ¹⁴C record (Muscheler et al., 2004). This uncertainty can theoretically be accounted for by calculating ¹⁴C production rates using a carbon cycle model. However, changes in Earth's carbon reservoirs are difficult to assess (Köhler et al., 2006). ¹⁰Be in mid-latitude regions is nearly exclusively scavenged from the atmosphere by precipitation (Heikkilä et al., 2013). Varying atmospheric circulation and scavenging during the about one month long tropospheric residence time (about 1 year stratospheric residence time) result in spatially non-uniform ¹⁰Be deposition patterns (Aldahan et al., 2008; Raisbeck et al., 1981). Despite these non-production effects, common changes in ¹⁰Be and ¹⁴C records are considered to reflect the cosmogenic radionuclide production signal, due to their common production mechanism and different chemical behavior (Lal and Peters, 1967; Muscheler et al., 2016).

To date, synchronization studies based on cosmogenic radionuclides are mainly limited to ¹⁴C records from trees and ¹⁰Be time-series from Arctic and Antarctic ice cores (Raisbeck et al., 2017; Muscheler et al., 2014). For example, Adolphi and Muscheler (2016) synchronized the Greenland ice core and IntCal13 time-scales for the last 11000 years. Synchronizing ¹⁰Be records in sedimentary archives opens the opportunity for the synchronization of paleoclimate records around the globe. Thereby, the temporal resolution of this approach is limited by the lowest resolution record involved. First studies underline the potential of varved lake sediments for recording the ¹⁰Be

production signal, down to annual resolution (Berggren et al., 2013, 2010, Czymzik et al., 2015, 2016a; Martin-Puertas et al., 2012).

In the following, we attempt to synchronize ¹⁰Be records from varved sediments of Lakes Tiefer See (TSK) and Czechowskie (JC) covering the grand solar minima at 250 a BP (Maunder Minimum), 2700 a BP (Homeric Minimum) and 5500 a BP with ¹⁴C production rates inferred from the IntCal13 calibration curve (Muscheler et al., 2014; Reimer et al., 2013). Annual ¹⁰Be time-series from both lake sediment archives yield the broad preservation of the ¹⁰Be production signal during solar cycles 22 and 23 (Czymzik et al., 2015). The targeted three grand solar minima comprise among the lowest solar activity levels throughout the last 6000 years (Steinhilber et al., 2012).

2. Study sites

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TSK (53°35'N, 12°31'E, 62 m a.s.l.) and JC (53°52'N, 18°14'E, 108 m a.s.l.) are situated within the Pomeranian Terminal Moraine in the southern Baltic lowlands (Fig. 1) (Dräger et al., 2017; Ott et al., 2016; Słowiński et al., 2017). The lake basins are part of subglacial channel systems formed at the end of the last glaciation and had no major inflows during the Holocene (Dräger et al., 2017; Ott et al., 2016). Both lakes are of similar size (TSK: 0.75 km²/JC: 0.73 km²), but the catchment of JC (19.7 km²) is about 4 times larger than that of TSK (5.5 km²) (Fig. 1). TSK sediments during the investigated grand solar minima are composed of alternating intervals of organic-, calcite-and rhodochrosite varves as well as intercalated non-varved sections (Dräger et al., 2017). JC sediments for these time windows comprise endogenic calcite varves with couplets of calcite and organic/diatom sub-layers, and an additional layer of resuspended littoral material since about 2800 a BP (Ott et al., 2016; Wulf et al., 2013). TSK and JC are located at the interface of maritime westerly and continental airflow. Mean annual precipitation is similar at both sites, 640 mm a⁻¹ at TSK and 680 mm a⁻¹ at JC (Czymzik et al., 2015).

3. Methods

3.1. Sediment sub-sampling and proxy records

25 Continuous series of sediment samples at ~20-year resolution (about 20 mm sediment) were extracted for ¹⁰Be measurements from sediment cores TSK11 and JC-M2015, based on varve chronologies (Dräger et al., 2017; Ott et al., 2016). Complementary sediment accumulation rate (SAR), geochemical X-ray fluorescence (μ-XRF) and total organic carbon (TOC) time-series were constructed using existing high-resolution datasets from the same sediment cores by calculating ¹⁰Be sample averages (Dräger et al., 2017; Ott et al., 2016; Wulf et al., 2016). Measured μ-XRF data (cps) were normalized by dividing by the sum of all elements, to reduce the effects of varying sediment properties (Weltje and Tjallingii, 2008).

3.2. ¹⁰Be extraction and AMS measurements

After spiking with 0.5 mg 9 Be carrier, authigenic Be was leached from 0.2 g ground sediment samples overnight with 8 M HCl at 60°C (Berggren et al., 2010). The resulting solutions were filtered to separate the undissolved fractions. Further addition of NH₃ and H₂SO₄ caused the precipitation of metal hydroxides and silicates, which were again removed by filtering. The remaining solutions were treated with EDTA to separate other metals and, then, passed through hydrogen form ion exchange columns in which Be was retained. Be was extracted from the columns using 4 M HCl and Be(OH)₂ precipitated through the addition of NH₃ at pH 10. The samples were washed and dehydrated three times by centrifuging and oxidized to BeO at 600°C in a muffle furnace. After mixing with Nb, AMS measurements of BeO were performed at the Tandem Laboratory of Uppsala University. Final 10 Be concentrations were calculated from measured 10 Be/ 9 Be ratios, normalized to the NIST SRM 4325 reference standard (10 Be/ 9 Be = 2.68 x 10 ${}^{-11}$) (Berggren et al., 2010).

3.3 Original chronologies

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The age models for TSK and JC sediments were constructed using a multiple-dating approach. Microscopic varve counts were carried out for both lake sediments. Non-varved intervals in TSK sediments were bridged based on varved thickness measurements in neighboring well-varved sediment sections. Independent age control for the TSK and JC varve chronologies was provided by radiocarbon dating and tephrochronology (for details see: Dräger et al., 2016, Ott et al., 2016, 2017; Wulf et al., 2013). Resulting chronological uncertainties are \pm 17 (TSK) and \pm 4 years (JC) for the Maunder Minimum, \pm 139 (TSK) and \pm 29 years (JC) for the Homeric Minimum as well as \pm 74 (TSK) and \pm 56 years (JC) for the 5500 a BP grand solar minimum (see Fig. 6).

3.4. Time-scale synchronization

Lag-correlation analyses were applied to determine best fits between the ¹⁰Be records from TSK and JC for the Maunder-, Homeric- and 5500 a BP grand solar minima and ¹⁴C production rates inferred from the IntCal13 calibration curve (Muscheler et al., 2014; Reimer et al., 2013). Before the correlation, all time-series were 75 to 500-year band-pass filtered and normalized by dividing by the mean, to reduce noise and increase the comparability (Adolphi et al., 2014). Significance levels for all correlation coefficients were calculated using 10000 iterations of a non-parametric random phase test, taking into account autocorrelation and trend present in the time-series (Ebisuzaki, 1997). Chronological uncertainty ranges were reported as the time-spans in which significances of correlations are below the given significant level. Before the analyses, all time-series were resampled to a 20-year resolution.

4. Results

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 10 Be concentrations (10 Be_{con}) were measured in 78 sediment samples from TSK and 73 sediment samples from JC (Figs. 2, 3 and S1). 10 Be_{con} in TSK sediments range from 1.13 to 7.09 x 10^8 atoms g^{-1} , with a mean of 3.91 x 10^8 atoms g^{-1} (Figs. 2 and S1). 10 Be_{con} in JC sediments vary between 0.93 to 3.82 x 10^8 atoms g^{-1} , around a mean of 1.89 x 10^8 atoms g^{-1} (Figs. 3 and S1). Mean AMS measurement uncertainties are 0.12 x 10^8 atoms g^{-1} for TSK and 0.07 x 10^8 atoms g^{-1} for JC samples (Figs. 2, 3 and S1). Due to the 1.387 ± 0.012 Ma long half-life of 10 Be (Korschinek et al., 2010), the effect of radioactive decay is negligible in our 10 Be records.

5. Discussion

10 5.1. ¹⁰Be production signal in TSK and JC sediments

Environment and catchment conditions can add non-production variations to $^{10}\text{Be}_{\text{con}}$ records from varved lake sediments (Berggren et al., 2010; Czymzik et al., 2015). In the following chapter we will, first, describe our approach used for detecting and correcting possible non-production features in our ^{10}Be time-series and, then, discuss possible mechanisms behind the statistically inferred connections.

To detect and reduce non-production effects in our ¹⁰Be time-series, we perform a three-step statistical procedure following Czymzik et al. (2016a), with a slight modification. First, multi-linear regressions were calculated between the ¹⁰Be_{con} records and TOC, SAR, Ca, Si, Ti proxy time-series from TSK and JC, reflecting changes in sediment accumulation and composition (Dräger et al., 2017; Ott et al., 2016; Wulf et al., 2016), to estimate the possible environmental influence (¹⁰Be_{bias}). Only the TOC and Ca time-series with significant contributions (p<0.1) for TSK and JC were included to the final multi-regressions. Subsequently, the resulting ¹⁰Be_{bias} time-series from TSK and JC sediments were subtracted from the original ¹⁰Be_{con} records in an attempt to construct an environment-corrected version of the ¹⁰Be record (¹⁰Be_{environment}). However, this statistical approach also removes variability in the ¹⁰Be_{con} records only coincident with variations in proxy time-series, but without a mechanistic linkage, potentially resulting in an overcorrection. Such coinciding variability can be introduced by solar activity variations causing ¹⁰Be production changes and climate variations imprinted in the proxy time-series. Therefore, final ¹⁰Be composite records (¹⁰Be_{comp}) were calculated by averaging the ¹⁰Be_{con} and ¹⁰Be_{environment} records from each site. To enhance the robustness of the corrections, the procedure was performed on the complete ¹⁰Be_{con} records from TSK and JC covering all three grand solar minima. Uncertainty ranges of the calculated ¹⁰Be_{comp} records are expressed as the differences between the ¹⁰Be_{con} and ¹⁰Be_{environment} time-series (Fig. 4).

Calculated ¹⁰Be_{comp} time-series from TSK and JC sediments yield modified trends, but similar multi-decadal variability as the original ¹⁰Be_{con} records during the Maunder- (TSK: r=0.84, p<0.01; JC: r=0.91; p<0.01), Homeric- (TSK: r=0.81, p<0.01; JC: r=0.74; p<0.01) and 5500 a BP grand solar minimum (TSK: r=0.89, p<0.01; JC: r=0.68; p<0.01) (Fig. 4). These linkages suggest that our correction procedure predominantly reduced trends in the ¹⁰Be_{con} records introduced by varying sedimentary TOC and Ca contents, but largely preserved multi-decadal variations

connected with varying ¹⁰Be production (Figs. 2, 3 and 4). Comparable linkages between measured and corrected ¹⁰Be records (based on a similar approach) were found in Lake Meerfelder Maar sediments covering the Lateglacial-Holocene transition as well as in recent TSK and JC sediments (Czymzik et al., 2015, 2016a).

The statistical connections to TOC and Ca for TSK and JC might point to depositional mechanisms of ¹⁰Be in lake sediment records. Significant contributions to the multi-regression as well as significant positive correlations for TSK (r=0.62, p<0.01) and JC (r=0.77, p<0.01) suggest a preferential binding of ¹⁰Be to organic material (Figs. 2 and 3). This result is supported by significant positive correlations of ¹⁰Be with TOC in two annually resolved timeseries from varved sediments of TSK and JC spanning solar cycles 22 and 23 and in Meerfelder Maar sediments covering the Lateglacial-Holocene transition (Czymzik et al., 2015, 2016a).

Significant contributions of Ca to the multi-regressions as well as significant negative correlations with ¹⁰Be for TSK (r=-0.68, p<0.01) and JC (r=-0.62, p<0.01) might point to a reduced affinity of ¹⁰Be for Ca (Figs. 2 and 3). A similar behavior was detected in studies about ¹⁰Be scavenging from the marine realm (Aldahan and Possnert, 1998; Chase et al., 2002, Simon et al., 2016).

5.2. ¹⁰Be_{comp} and group sunspot numbers

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To evaluate the preservation of the cosmogenic radionuclide production signal based on observational data, the ¹⁰Be_{comp} time-series from TSK and JC were compared with a group sunspot number record reaching back to 340 a BP (AD 1610) (Svalgaard and Schatten, 2016) (Fig. 5). Since sunspot and cosmogenic radionuclide records reflect different components of the heliomagnetic field (closed and open magnetic flux) no perfect correlation is expected (Muscheler et al., 2016). Nevertheless, a comparison of a ¹⁴C based solar activity reconstruction with group sunspot data points to a largely linear relationship between both types of data (Muscheler et al., 2016).

Variations in the ¹⁰Be_{comp} records from TSK and JC resemble multi-decadal to centennial variability in the group sunspot number time-series with highest values around the Maunder Minimum (Fig. 5). Secondary ¹⁰Be_{comp} maxima in TSK and JC sediments broadly coincide with the Dalton Minimum and solar activity minimum around 30 a BP (AD 1920) (Fig. 5). In JC sediments, the ¹⁰Be_{comp} excursion from -50 to 0 a BP (AD 2000-1950) without an expression in the group sunspot number record as well as the about 20-year delayed Maunder Minimum response could be explained by transport of 'old' ¹⁰Be from the littoral to the coring site (see details on the sub-layer of resuspended littoral sediments in JC varves back to 2800 a BP in Section 5.3) and/or spatially inhomogeneous ¹⁰Be deposition patterns (Fig. 5).

5.3. Synchronizing TSK/JC ¹⁰Be with IntCal13 ¹⁴C

Shared variance of ¹⁰Be and ¹⁴C records can be interpreted in terms common changes in cosmogenic radionuclide production (Czymzik et al., 2016a; Muscheler et al., 2014). Moreover, it provides the opportunity to synchronize cosmogenic radionuclide records from different archives (Adolphi and Muscheler, 2016). Lag-correlation analyses were performed to synchronize the TSK and JC ¹⁰Be_{comp} records covering the Maunder-, Homeric- and 5500 a BP grand solar minima with ¹⁴C production rates from the IntCal13 calibration curve (Muscheler et al., 2014).

Best fits with IntCal13 14 C production rates were obtained, when the 10 Be_{comp} records from TSK were shifted by 8 - 12/+4 years (Maunder Minimum; r=0.47, p<0.1), 31 -16/+12 years (Homeric Minimum; r=0.68, p<0.01) and 86 - 22/+18 years (5500 a BP grand solar minimum; r=0.37, p<0.05) towards the past (Fig. 6). All three best fits occur within the given chronological uncertainties of \pm 17 years (Maunder Minimum), \pm 139 years (Homeric Minimum) and \pm 74 years (5500 a BP grand solar minimum) (Fig. 6). The on average <10-year resolution of the varve-based sedimentation rate chronology for TSK sediments around the Maunder Minimum due to non-varved intervals, does not affect our analyses conducted on records at 20-year resolution.

The best fit between the ¹⁰Be_{comp} record from JC sediments during the 5500 a BP grand solar minimum and IntCal13 ¹⁴C production rates (r=0.81, p<0.01) was determined, when the ¹⁰Be record was shifted for 29 -8/+7 years towards present (within the given chronological uncertainty of ±56 years) (Fig. 6). No significant correlations between the ¹⁰Be_{comp} records from JC and ¹⁴C production rates were obtained for the Maunder- and Homeric Minima, within the respective varve counting uncertainties of ±4 and ±29 years (Fig. 6). This lack of significant correlation might be explained by a change in sedimentation at about 2800 a BP (Fig. 6). Since that time JC varves include an additional sub-layer of littoral calcite and diatoms transported to the profundal by wave driven water turbulences in fall. Presumably, the resuspended material also contains 'old' ¹⁰Be inhibiting the clear detection of the expected ¹⁰Be production signal (Fig. 6). Since the ¹⁰Be signal present in the resuspended sediments is unknown, this uncertainty is difficult to correct for. Comparable influences of sediment resuspension were also found in a sample from an annually resolved ¹⁰Be record from JC sediments covering the period AD 2009-1988 (Czymzik et al., 2015). A varve with an exceptionally thick (3.7 mm) layer of resuspended littoral diatoms and calcite deposited in fall 2003 reveals anomalous ¹⁰Be concentrations (Czymzik et al., 2015).

6. Conclusions

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Detecting and aligning the common cosmogenic radionuclide production variations allows the synchronization of ¹⁰Be time-series from TSK sediments covering the Maunder-, Homeric- and 5500 a BP grand solar minima and JC sediments for the 5500 a BP grand solar minimum to IntCal13 ¹⁴C production rates. These synchronizations provide a novel type of time-marker for varved lake sediment archives enabling improved chronologies and robust investigations of proxy responses to climate variations. Mismatches between ¹⁰Be in JC sediments and ¹⁴C production rates during the Maunder- and Homeric Minima are likely associated with in-lake resuspension of 'old' ¹⁰Be, altering the expected ¹⁰Be production signal.

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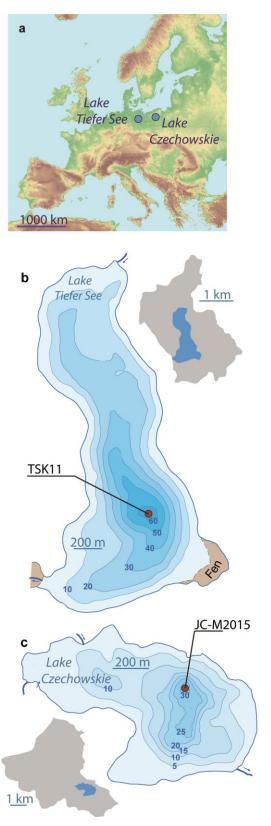


Figure 1. Settings of Lakes Tiefer See (TSK) and Czechowskie (JC). (a) Location of TSK and JC in the southern Baltic lowlands. (b) Bathymetric map of TSK with position of sediment core TSK11 and lake-catchment sketch. (c) Bathymetric map of JC with position of sediment core JC-M2015 and lake-catchment sketch.

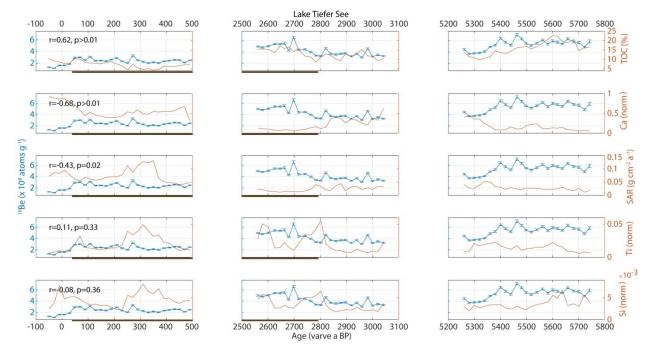


Figure 2. ¹⁰Be concentrations (¹⁰Be_{con}) in Lake Tiefer See (TSK) sediments around the Maunder-, Homeric- and 5500 a BP grand solar minima and corresponding proxy time-series from the same archive. ¹⁰Be_{con} compared with sediment accumulation rates (SAR), total organic carbon (TOC), Ti, Ca and Si. Correlation coefficients were calculated for the complete time-series covering all three grand solar minima. Significance levels of correlations were calculated using 10000 iterations of a non-parametric random phase test taking into account trend and autocorrelation present in the time-series (Ebisuzaki, 1997). Error bars indicate AMS measurement uncertainties. Non-varved intervals in TSK sediments are indicated by bars.

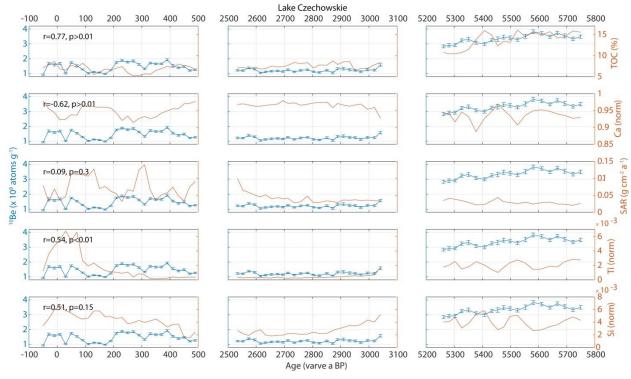


Figure 3. ¹⁰Be concentrations (¹⁰Be_{con}) in Lake Czechowskie (JC) sediments around the Maunder-, Homeric- and 5500 a BP grand solar minima and corresponding proxy time-series from the same archive. ¹⁰Be_{con} compared with sediment accumulation rates (SAR), total organic carbon (TOC), Ti, Ca and Si. Correlation coefficients were calculated for the complete time-series covering all three grand solar minima. Significance levels of correlations were calculated using 10000 iterations of a non-parametric random phase test taking into account trend and autocorrelation present in the time-series (Ebisuzaki, 1997). Error bars indicate AMS measurement uncertainties.

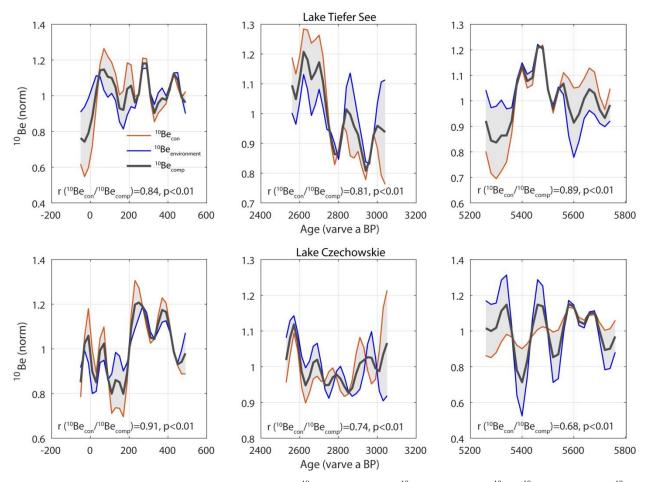


Figure 4. Lakes Tiefer See (TSK) and Czechowskie (JC) ¹⁰Be concentration (¹⁰Be_{con}), corrected ¹⁰Be (¹⁰Be_{environment}) and ¹⁰Be composite (¹⁰Be_{comp}) time-series around the Maunder-, Homeric- and 5500 a BP grand solar minima. All time-series are resampled to a 20-year resolution and normalized by dividing by the mean. A 75-year low pass filtered was applied to reduce noise. Uncertainty ranges of ¹⁰Be_{comp} (gray shadings) are expressed as the differences between the ¹⁰Be_{con} and ¹⁰Be_{environment} time-series. Significance levels of correlations between ¹⁰Be_{con} and ¹⁰Be_{comp} were calculated using a random phase test (Ebisuzaki, 1997).

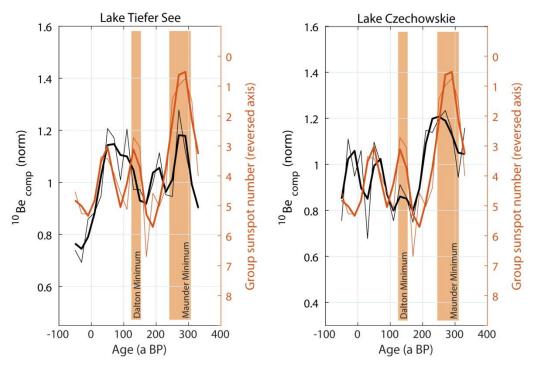
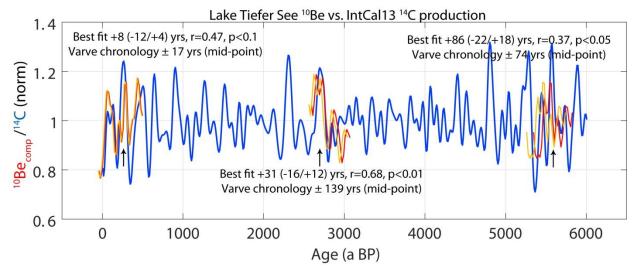


Figure 5. ¹⁰Be composites (¹⁰Be_{comp}) from Lakes Tiefer See (TSK) and Czechowskie (JC) compared with group sunspot numbers back to 340 a BP (Svalgaard and Schatten, 2016). Time-windows of the Maunder and Dalton solar minima are highlighted (Eddy, 1976; Frick et al., 1997). Time-series are shown at 20-year resolution (thin lines) and with a 75-year low-pass filter, to reduce noise (thick lines). ¹⁰Be_{comp} records are normalized by dividing by the mean.



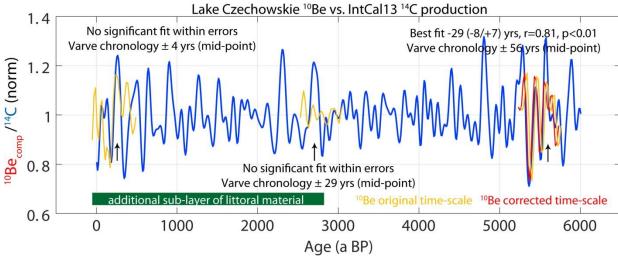
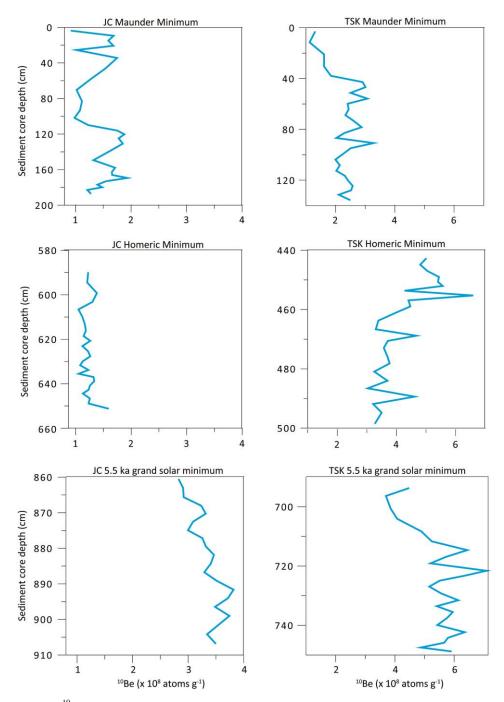


Figure 6. Synchronization of ¹⁰Be composites (¹⁰Be_{comp}) from Lakes Tiefer See (TSK) and Czechowskie (JC) for the Maunder-, Homeric- and 5500 a BP grand solar minima with ¹⁴C production rates from the IntCal13 calibration curve (Muscheler et al., 2014). Best fits between the records were calculated using lag-correlation, based on given chronological uncertainties (Dräger et al., 2017; Ott et al., 2016). Significance levels of the correlations were calculated using 10000 iterations of a non-parametric random phase test taking into account autocorrelation and trend present in the time-series (Ebisuzaki, 1997). Uncertainies are given as the time-spans in which the significances of the correlations are below their respective significant levels. ¹⁰Be_{comp} is shown on its original time-scale and, if applicable, synchronized with IntCal13 ¹⁴C production rates. An interval with an additional sub-layer of resuspended littoral material in JC varves is indicated. Arrows mark the peaks of the targeted three grand solar minima.



Supplementary Fig S1. ¹⁰Be concentrations in Lakes Tiefer See (TSK) and Czechowskie (JC) sediments around the Maunder-, Homeric- and 5.5 ka grand solar minima plotted against sediment core depth.