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The Holocene

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Comparison of Varve and ^{14}C Chronologies from Steel Lake, Minnesota, USA

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A **Abstract:** Annually laminated sediments (varves) offer an effective means of acquiring high-quality paleoenvironmental records. However, the strength of a varve chronology can be compromised by a number of factors, such as missing varves, ambiguous laminations, and human counting error. We assess the quality of a varve chronology for the last three millennia from Steel Lake, Minnesota, through comparisons with nine AMS ^{14}C dates on terrestrial plant macrofossils from the same core. These comparisons revealed an overall 8.4% discrepancy, primarily because of missing/uncountable varves within two stratigraphic intervals characterized by low carbonate concentrations and obscure laminations. Application of appropriate correction factors to these two intervals results in excellent agreement between the varve and ^{14}C chronologies. These results, together with other varve studies, demonstrate that an independent age-determination method, such as ^{14}C dating, is usually necessary to verify, and potentially correct, varve chronologies.

A **Key words:** varve chronology, ^{14}C chronology, laminated sediment, counting error, varve-count adjustment, paleoclimate.

A Introduction

Varved sediments play an important role in paleoenvironmental studies because of their potential to offer calendar-year-resolution chronologies (e.g., O'Sullivan, 1983; Hughen et al., 2000). Such sediments experience minimal disturbance after deposition, enabling the calibration of environmental proxies with instrumental or historical data (e.g., Lotter and Birks, 1997; Wehrli et al., 1997) and high-temporal-resolution environmental reconstructions (e.g., Goslar et al., 1990; Rittenour et al., 2000). Over the past several decades, varves have provided chronological frameworks for a number of paleoclimatic, paleoceanographic, and paleoecologic studies (e.g., Craig, 1972; Anderson et al., 1993; Hu et al., 1999; von Rad et al., 1999; Goslar et al., 2000; Tiljander et al., 2003). However, the accuracy of a varve-based chronology is susceptible to various sources of error, including discontinuous varve deposition through time, obscure laminations, and human counting error. For example, cumulative errors of varve counting are often greater than 10% (e.g., Sprowl, 1993; Aardsma, 1996), potentially jeopardizing the reliability of paleo-studies that require determination of precise timing.

As part of a project with an overall goal of high-temporal-resolution environmental reconstructions, we have analyzed varved sediments from Steel Lake (46°58' N, 94°41' W; 415.4 m a.s.l.; Fig. 1a), Minnesota, for multiple proxy indicators (e.g., carbon and oxygen isotopes, varve thickness, and elemental composition) at annual to sub-decadal resolution. The reliability of the paleoenvironmental inferences ultimately depends on the quality of our varve chronology. The goals of this paper are to evaluate the Steel Lake

varve chronology for the past three millennia and to explore error-correction methods through comparison of the varve chronology with accelerator-mass-spectrometry (AMS) ^{14}C dates on terrestrial plant macrofossils. Results show that substantial varve chronological errors exist within stratigraphic intervals of obscure laminations characterized by low carbonate contents. In conjunction with other varve records (e.g., Sprowl, 1993; Hajdas et al., 1995a; Hajdas et al., 1995b; Aardsma, 1996; Brauer et al., 2000), these results underscore the importance of verifying varve counts with an independent chronology.

A **Study site and methods**

Steel Lake is located within the Itasca moraine in north-central Minnesota (Mooers and Norton, 1997; Wright et al., 2004). The lake formed as stagnant ice blocks melted (Wright, 1993), and it is surrounded by thick sandy glacial deposits (Winter and Rosenberry, 1997) with a watershed area of 1.08 km^2 . The relatively small surface area (0.23 km^2), great water depth ($\sim 21 \text{ m}$, Fig. 1b), and anoxic bottom water of the lake (Fig. 1c) facilitate the preservation of seasonal depositional patterns (O'Sullivan, 1983).

Two freeze cores containing the sediment-water interface were recovered with a mixture of dry ice and ethanol (Wright, 1980) during the summer of 2001. For deeper sediments, three stratigraphically overlapping cores were retrieved with a modified piston corer (Wright et al., 1984). In order to cover potential gaps between the freeze and piston cores, two additional short cores were taken with polycarbonate tubes. All of the cores

were collected from the deepest part of the lake (Fig. 1b). The cores were easily matched by lamination patterns to provide a continuous sedimentary record.

Petrographic thin sections were prepared from an entire freeze core and from selected intervals of a piston core for detailed examination of microscopic varve structures (Camuti and McGuire, 1999; Lotter and Lemcke, 1999). Varve counting proceeded from the core top downward, with the first complete lamina deposited in the year 2000. Counting was conducted independently by two people, both on the sediment cores and on the core images, to assess counting errors.

In order to verify the varve counts of the uppermost sediment, radiocesium (^{137}Cs) dating was performed on continuous subsamples from a freeze core for the period of 1946–1975 (based on varve count). Each subsample consisted of three dark-light couplets. The subsamples were submitted to the St. Croix Watershed Research Station, Minnesota, for ^{137}Cs analysis. ^{137}Cs activity was measured with an Ortec[®] high-purity Germanium photon detector coupled to a digital gamma-ray spectrometer.

To concentrate terrestrial plant macrofossils for AMS ^{14}C dating, we sliced one half of a sediment core into contiguous 2-cm segments and washed them through a 250- μm -mesh sieve. The samples selected for ^{14}C analysis were pretreated with an acid-base-acid protocol at the University of Illinois and submitted to the Center for Accelerator Mass Spectrometry at Lawrence Livermore National Laboratory. To compare with the varve chronology, we converted ^{14}C dates individually to calibrated ages using CALIB

v4.4html (<http://radiocarbon.pa.qub.ac.uk/calib/>, Stuiver and Reimer, 1993) and the decadal atmospheric calibration data set (Stuiver et al., 1998). The median-probability values of the calibrated ages were used to represent the ages of ^{14}C -dated stratigraphic levels. Both the calibrated ^{14}C ages and their 2σ ranges were expressed in calibrated years before 1950 AD (cal BP). In addition, we used the OxCal v3.9 calibration program (Ramsey, 1995; Ramsey, 2003) to evaluate the effectiveness of varve-chronology adjustments, derive the final varve chronology, and estimate errors.

A Results

Each varve is characterized by a couplet of a light layer and a dark layer (Fig. 2a). Petrographic thin sections reveal further details of rhythmic sedimentological variations related to seasonal changes (Nuhfer et al., 1993). In particular, each varve generally consists of three sublaminae: a layer of highly concentrated calcite ($\sim 1\ \mu\text{m}$ sized grains, Fig. 2b) and diatoms with scattered pollen grains deposited during the growing season; a thin layer of diatoms mixed with brownish iron-rich material representing the fall overturn; and a winter layer characterized by dark organic debris. The fall and winter layers are often indistinguishable by the naked eye. A total of 2872 varves were counted in the upper ~ 3.5 m of the cores (Table 1), below which were middle-Holocene sediments with ambiguous laminations and turbidites. Over a randomly chosen 1100-year interval, the counting difference between the two people is $\sim 2.2\%$. Varve thickness ranges from 0.46 to 3.88 mm with a mean of 1.22 mm for the period of interest. The varves are well defined except within two sections, between varve counts 312 and 632 and between

counts 2267 and 2417. These intervals are characterized by obscure laminations (Fig. 2a) with overall darker colors and average carbonate contents 10–15% lower than those of the adjacent segments containing similar numbers of varves.

The maximum ^{137}Cs deposition, which resulted from atmospheric nuclear weapons testing, occurred during 1963–64 (Jaakkola et al., 1983; Erten, 1987; Holmes, 1998). The ^{137}Cs activity at Steel Lake has a peak value of 4.420 pCi/g for the varve-dated period of 1964–1966 (Table 2), which verifies our varve counts back to the 1960s within the limit of sample resolution. These ^{137}Cs results, the high quality of the laminations, and the consistency of counts between people confirm the deposition of varves since 2000 AD and allow the direct association of the 50th (± 1) counted varve with 1950 AD. For convenient comparison with calibrated ^{14}C ages (in cal BP), varve counts are expressed on a varve BP scale hereafter in this paper, with 0 varve BP being associated with the 50th counted varve and 1950 AD (0 cal BP).

Nine AMS ^{14}C dates were obtained within the varve-counted section (Table 1). All dated plant macrofossils were well preserved, suggesting that they had short residence times in the watershed before being incorporated in the sediment. The relatively large analytical errors of some of the ^{14}C dates reflect the influence of sample-size-dependent background corrections. The median-probability values of the calibrated ^{14}C ages (Table 1, column 4) are in stratigraphic order and span the last 3 ka cal BP with separations between ages of 190–900 years (Table 1, column 5).

A Discussion

B *Comparison of ^{14}C ages and varve counts*

A The calibrated- ^{14}C and varve ages should show an overall 1:1 relationship if the two chronologies agree. Instead, weighted linear regression of our data yields a slope of 1.084 ± 0.019 , which is significantly greater than 1.0 at the 99% confidence level (Fig. 3a). This slope suggests an 8.4% discrepancy between our varve and calibrated- ^{14}C chronologies. Three possibilities may account for this discrepancy: 1) the varve counts are accurate and the calibrated ^{14}C dates are older than the true sediment ages; 2) the calibrated ^{14}C dates are accurate and the varve counts underestimate the true sediment ages; or 3) both the varve counts and the calibrated ^{14}C dates have errors.

Because all of our terrestrial plant macrofossils for ^{14}C analysis were well preserved, it is unlikely that these materials were subject to reservoir aging effects that would have caused them to be systematically older than the true sediment ages. In addition, the difference between the calibrated- ^{14}C and varve chronologies generally increases with age (Figs. 3a, 3b). This pattern points towards varve-count underestimation of the true sediment ages as the cause of the difference between the two chronologies.

To help pinpoint the cause of the discrepancy between the two chronologies, we compared the calibrated ^{14}C -age difference between each pair of the adjacent ^{14}C samples with the varve count between each ^{14}C -sample pair (Table 1, columns 5 and 7). This

comparison shows that the discrepancy is primarily caused by the large deviations of two data points representing the intervals between ^{14}C dates of 340 ± 40 BP and 780 ± 50 BP and between ^{14}C dates of 2160 ± 50 BP and 2490 ± 100 BP (Fig. 3c, Table 1, column 8). Removal of them reduces the regression slope from 1.041 to 0.997 (Fig. 3c), suggesting that the discrepancy between the two chronologies mainly occurs within these two intervals. These regression analyses may be biased because they appear to be greatly influenced by a single data point representing the greatest age differences (Table 1, row 3 and columns 5 and 7). However, the removal of this point from regression does not alter our conclusion.

If a single ^{14}C date departs from the true age (e.g., is older), a pair of differences of opposite signs are expected, with one above the 1:1 line and the other of a similar offset below the 1:1 line. However, neither of the two extreme age differences discussed above has a comparable age difference of opposite sign (Table 1, column 8). This pattern suggests that the deviations of these two points from the 1:1 line are not caused by erroneous ^{14}C dates. Instead, localized errors in the varve chronology are the likely cause of the deviations. Such a varve-age error would affect only one data point in an age-difference plot (Fig. 3c), and the cumulative counting error would propagate downward to all levels below (Fig. 3a, 3b).

The sections between ^{14}C dates of 340 ± 40 BP and 780 ± 50 BP (equivalent to 364 and 564 varve BP; Table 1) and between ^{14}C dates of 2160 ± 50 BP and 2490 ± 100 BP (equivalent to 2097 and 2357 varve BP; Table 1) roughly correspond to the two low-

carbonate zones between 262 and 582 varve BP and between 2217 and 2357 varve BP, respectively. The low content of carbonate, which constitutes a major component of the light sub-lamina that is important for varve delineation, probably has impeded accurate varve counting within these intervals. It is also possible that partial carbonate dissolution occurred during and after deposition (Ohlendorf and Sturm, 2001; Müller et al., 2003), resulting in missing varves in these intervals. We cannot determine the relative importance of these two factors.

The presence of substantial errors in the Steel Lake varve chronology is consistent with the results of varve analyses at other sites where varve chronologies can be evaluated with independent dating controls. For example, excellent varve sequences are available from Lake Meerfelder Maar (Brauer et al., 2000) and Lake Holzmaar (Hajdas et al., 1995b; Hajdas and Bonani, 2000), Germany. As at Steel Lake, each of these varve chronologies has a corresponding chronology based on multiple ^{14}C dates. For the same late-Holocene period, most of the data points from these two lakes (Fig. 4a) deviate substantially from the ^{14}C calibration curve (Stuiver et al., 1998). The best linear-regression fits to the varve and calibrated- ^{14}C chronologies at these two sites reveal departures from the 1:1 relationship with the magnitudes of discrepancy between the two chronologies comparable with that at Steel Lake.

B *Varve chronological adjustments*

Two approaches can be used to correct errors in varve chronologies caused by missing/uncountable varves. The first is to apply a uniform-correction factor, which is derived from the linear-regression slope of calibrated ^{14}C -ages and varve counts, to the entire varve chronology. This simple adjustment is appropriate when the varve quality does not vary greatly, such that the counting error is approximately constant throughout the varve record. However, the uniform correction may add error to accurately counted sections if the varve quality, and hence varve-counting reliability, varies between different segments of the core.

An alternative approach is to examine varve counts and ^{14}C ages by intervals (e.g., between adjacent ^{14}C samples) and apply corrections only to the intervals where adjustments in varve counts are warranted (local correction). There are two ways of applying a local correction: inserting an appropriate number of years at a particular point if evidence of erosion or a depositional hiatus exists, or using a constant adjustment-factor throughout the interval of discrepancy. The local-correction approach is more appropriate than the uniform correction when sedimentological evidence shows that specific intervals of the varve sequence are problematic. This approach has been used in previous varve studies. For example, 878 years were inserted to the Lake Holzmaar varve chronology for sediment below 500 cm, where a 24-cm sediment interval had no diatom-rich sublaminae (Hajdas et al., 1995b). At Soppensee Lake, Switzerland, an estimate of 550 years was added to the varve chronology to compensate for the unlaminated segments during the Younger Dryas (Hajdas and Bonani, 2000). Similarly, ~600 years were added to the varve chronology for the disturbed top sediment at Lake of

the Clouds, Minnesota (Stuiver, 1971). In all of these cases, the varve chronological adjustments greatly improved the agreement between varve and ^{14}C chronologies.

We applied both the uniform and local corrections to the Steel Lake varve chronology to test these approaches. For the uniform correction, we multiplied our original varve counts by the linear-regression slope of 1.084 (Fig. 3a) such that the overall pattern of our calibrated ^{14}C dates/varve counts fell on the 1:1 line. To further evaluate the relationship between the ^{14}C dates and the results of this uniform correction with respect to the ^{14}C calibration curve (Stuiver et al., 1998), we used the V_sequence analysis of the OxCal v3.9 calibration program (Ramsey et al., 2001). This analysis incorporates ^{14}C dates, varve counts (to define the age gap between adjacent ^{14}C dates), and their uncertainties (gap error is approximated as $1.6\% [= (2.2\% / 2) \times \sqrt{2}]$, where 2.2% is the counting difference between the two people] of the varve counts) to evaluate the agreement based on a Bayesian probabilistic approach. The results of the V_sequence analysis (Fig. 5a) showed a low agreement index of 41.7% between our two chronologies, with a significant disagreement between the calibration curve and the 780 ± 50 BP date at the uniform-correction age of 590 varve BP. This poor overall agreement indicates that the uniform correction approach did not provide an appropriate correction for the errors in the Steel Lake varve chronology. This conclusion is not surprising because, as discussed above, the discrepancies between the two chronologies appear to be introduced mainly within the two low-carbonate segments of the core (i.e., 262 to 582 varve BP and 2217 to 2357 varve BP).

For a local correction, we applied a constant factor throughout each of the two problematic segments because no evidence exists for a single erosion layer or a hiatus in either of these intervals. Specifically, we added 120 and 115 years to the intervals between 262 and 582 varve BP and between 2217 and 2357 varve BP, respectively. The selection of 120 and 115 years was based on comparisons between the original varve counts and calibrated ^{14}C ages (Table 1, column 8) and on linear regression of the adjusted varve counts and the calibrated ^{14}C ages. When plotted against this locally corrected varve chronology, the calibrated ^{14}C dates appeared to be largely consistent with the ^{14}C calibration curve (Fig. 4b). On the basis of the V_sequence analysis of this chronology, we derived the final varve chronology (Table 1, column 10), which shows good agreement with all of the ^{14}C dates, with an overall agreement index of 135.6% (Fig. 5b; Fig. 6). These results indicate that the local correction approach provided a statistically appropriate correction for the errors in the varve chronology of Steel Lake (Table 1, column 10; Fig. 6).

We estimated the uncertainty of the final varve chronology using the V_sequence analysis. The new error estimates reflect the cumulative uncertainty of varve counting and the constraints imposed by the ^{14}C dates. The resultant 2σ ranges (71 ± 20 , Table 1) are greatly reduced compared with those based on the calibrated ^{14}C -ages alone (340 ± 164 , Table 1).

A Conclusion

Our results emphasize that varve chronologies can potentially have large errors within certain stratigraphic intervals. The estimated cumulative error of about 240 years is seemingly trivial in the context of the time span of the entire record (~3000 years). It is also relatively small in comparison with the 2σ ranges of typical calibrated ^{14}C ages. However, the errors are large relative to the specific intervals involved, and they propagate downcore to affect much of the varve chronology. This study demonstrates that, with an adequate number of high-quality ^{14}C dates or other independent age controls (e.g., tephra of known ages), varve chronological errors can be identified and corrected, especially with the support of sedimentological evidence. The OxCal V-sequence analysis provides a statistically reliable basis for adjusting varve chronologies using ^{14}C dates and for estimating the associated errors.

These results do not imply that varve chronologies lack major advantages over ^{14}C dating. For example, the absolute durations of climatic and vegetational transitions, especially those at sub-decadal to centennial time scales, often cannot be determined with ^{14}C ages because of the varying sedimentation rates for the periods of interest (e.g., Fig. 3b) and because of the large age ranges usually resulting from ^{14}C dating and calibration. Varves can provide such sub-decadal to centennial information once they have been rigorously evaluated with independent chronological markers.

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Figure 1. a) Location of Steel Lake in Minnesota. **b)** Bathymetry (contour lines in meters) of Steel Lake. ‘X’ indicates the coring site. **c)** Dissolved oxygen (DO) profile at Steel Lake during the summer of 2002. Measurements were taken with a YSI® 85 DO & conductivity meter.

Figure 2. a) Varved sediments from Steel Lake. The upper image shows an example of distinct varves, and the lower image illustrates obscure varves. Both images have the same orientation and scale. **b)** Environmental scanning electron microscope (ESEM) image of Steel Lake sediment. The small grains are calcite crystals precipitated within the lake.

Figure 3. a) Comparison of the calibrated ^{14}C ages and the original varve chronology. The thick line represents the weighted linear fit to the data points (numbers are raw varve counts) with a regression slope of 1.084. The thin line represents the 1:1 relationship. **b)** Age-depth curves based on linear interpolation of the calibrated ^{14}C ages, on the original varve chronology, and on the locally corrected (LC) varve chronology. The divergence between age-depth curves based on the calibrated ^{14}C ages and the original varve counts indicates the cumulative discrepancy between the two chronologies, which is likely due to counting inaccuracy. The smaller differences between the LC varve chronology and the linear interpolation of the calibrated ^{14}C ages illustrate the inaccuracies inherent in the ^{14}C -age linear interpolation that are caused by variations in sedimentation rate; this highlights the advantage of a varve chronology at sub-decadal to centennial time scales. **c)**

Age differences between the adjacent calibrated ^{14}C ages vs. varve-count differences between adjacent ^{14}C -dated stratigraphic levels. The thick line is a linear-regression fit with a slope of 1.041, and the thin line is the 1:1 relationship. The two data points in the oval have the greatest deviation from both the linear-regression and 1:1 lines. The numbers next to the data points indicate the order of the age differences from the core top.

Figure 4. Comparison of ^{14}C and varve chronologies with the INTCAL 98 atmospheric ^{14}C calibration curve (thin wiggle line, Stuiver et al., 1998) as a reference. A left (or above) offset from the calibration curve indicates that either the ^{14}C date is too old if varve counts are accurate, or a varve undercount if the ^{14}C date is accurate. The opposite scenario is true for a right (or below) offset. **a)** Raw ^{14}C dates (w/ 2σ error bars) vs. varve counts from Lake Holzmaar (Germany) and Lake Meerfelder Maar (Germany). **b)** Raw ^{14}C dates (w/ 2σ error bars) vs. original varve counts (empty diamonds) and the LC varve chronology (solid triangles) from Steel Lake.

Figure 5. Comparisons of the calibrated ^{14}C ages and the adjusted varve chronologies. The lines are weighted linear-regression fits, and the numbers next to data points denote the agreement index following varve correction. The agreement index (A) was calculated as a pseudo-Bayes-Factor (Ramsey et al., 2001). The agreement is considered as poor when $A < 60\%$ (http://www.rlaha.ox.ac.uk/oxcal/oper_an.htm#prob_agree). **a)** Uniform correction with a constant correction factor of 1.084. The overall agreement is poor ($A = 41.7\%$). **b)** Local correction with varve adjustments confined within intervals between

raw varve counts of 262–582 varve BP, and 2217–2357 varve BP. The overall agreement is good ($A = 135.6\%$).

Figure 6. OxCal V_sequence result of the ^{14}C dates and the locally corrected varve chronology at Steel Lake. The labels inside the plot are CAMS numbers except for ‘C’ and ‘S’, which denote the combined ^{14}C date (1871 ± 31 BP) and the surface date (0 varve BP), respectively. The rectangular box overlaying the calibration curve indicates the dating error, with the inner and outer boxes as the 1σ and 2σ error ranges, respectively. The height of the box represents the raw ^{14}C dating error; and the width of the box represents the constrained error of the calibrated ^{14}C date and the locally adjusted varve chronology.

Table 1. AMS ^{14}C dates and varve counts, and comparison of varve counts and calibrated ^{14}C ages between adjacent ^{14}C samples from Steel Lake

CAMS #	Depth (cm)	$^{14}\text{C} \pm \text{Err}$ (BP)	Calib. ^{14}C Age (2 σ range) *	Diff. in ^{14}C Age ‡	VC ξ	Diff. in VC	Diff. btw VC & ^{14}C	VC-UC**	VC-LC** (2 σ range η)	Dated Material
66721	2183.7	340 \pm 40	390 (310, 490)	390	364	364	-26	380	400 (380, 420)	Bark
68535	2211	780 \pm 50	710 (650, 790)	320	564	200	-120	590	680 (660, 700)	Leaf + bark
71309	2330	1700 \pm 120	1610 (1350, 1870)	900	1472	908	8	1590	1600 (1560, 1630)	1 <i>Betula</i> (<i>Be</i>) seed
71310	2352	1880 \pm 40			1662					1 Cyperaceae (<i>Cy</i>) seed
71311	2356	1850 \pm 60			1702					1 <i>Be</i> + 1 <i>Cy</i> seeds
§	2353.9	1871 \pm 33	1800 (1720, 1880)	190	1682	210	20	1810	1800 (1770, 1850)	
68536	2392.5	2160 \pm 50	2160 (2000, 2310)	360	2097	415	55	2270	2220 (2180, 2260)	Charcoal flakes
68537	2411.5	2490 \pm 100	2560 (2350, 2760)	400	2357	260	-140	2550	2600 (2550, 2640)	1 Polygonaceae seed
68538	2428.6	2680 \pm 90	2800 (2490, 3000)	240	2562	205	-35	2770	2800 (2760, 2840)	2 <i>Betula/Alnus</i> seeds
71312	2448	2920 \pm 90	3070 (2850, 3340)	270	2767	205	-65	3000	3000 (2960, 3050)	1 <i>Alnus</i> seed

§ Pooled date from the two ^{14}C dates at depths of 2352 and 2356 cm, which are statistically indistinguishable. These two dates were

combined using CALIB v4.4 (Stuiver and Reimer, 1993) to yield a pooled mean, which was used in subsequent calculations. The

varve count corresponding to the pooled ^{14}C date is the mean of the varve counts corresponding to the original dates.

* Median-probability values of calibrated ^{14}C ages were chosen to represent the ages of ^{14}C -dated stratigraphic levels. The 2 σ ranges

encompass the >99% probability distributions of the calibrated ages.

‡ Age difference between adjacent ^{14}C samples was calculated from the median-probability value of the calibrated ^{14}C age.

ξ VC: varve count. Listed VC is the midpoint of the corresponding ^{14}C sample.

** UC : Uniform Correction; LC: Local Correction.

η The 2 σ range was estimated from the V_{sequence} analysis on both ^{14}C dates and LC varve chronology.

Table 2. ^{137}Cs dates from Steel Lake

Varve Age	^{137}Cs (pCi/g*)	S.D. (pCi/g)
1975-73	1.600	0.182
1972-70	1.610	0.174
1969-67	2.750	0.263
1966-64	4.420	0.247
1963-61	2.510	0.214
1960-58	1.790	0.134
1957-55	0.868	0.115
1954-52	0.484	0.113
1951-49	0.000	ND
1948-46	0.000	ND

* pCi/g - picocuries per gram.

Fig. 1

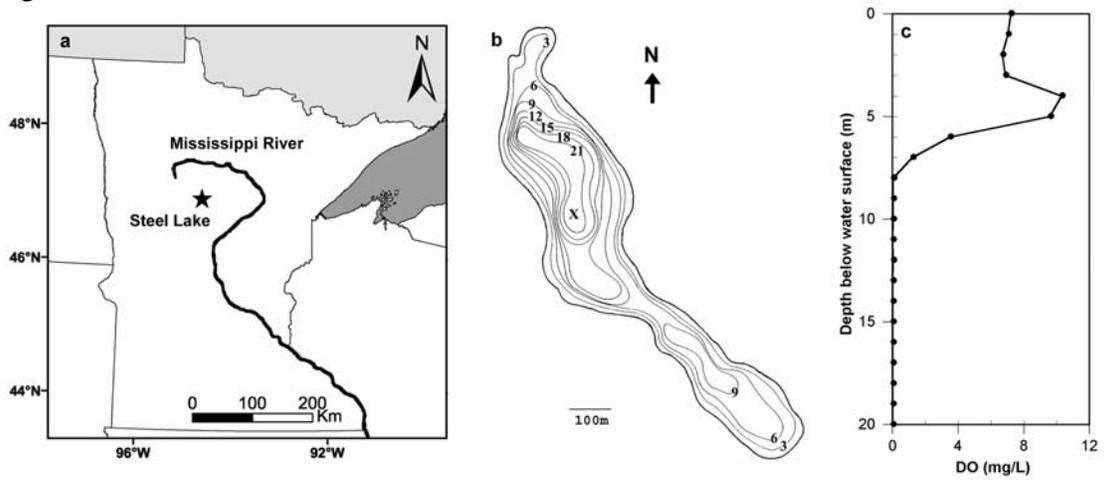
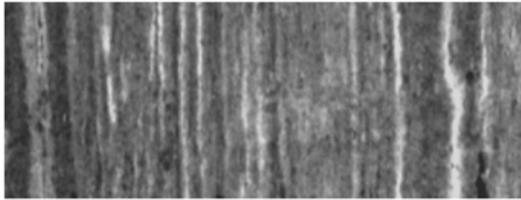
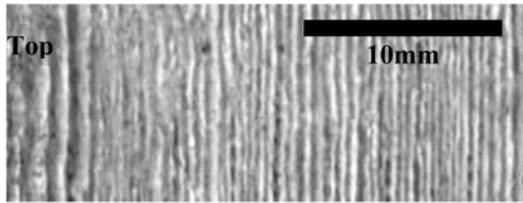


Fig. 2

a



b

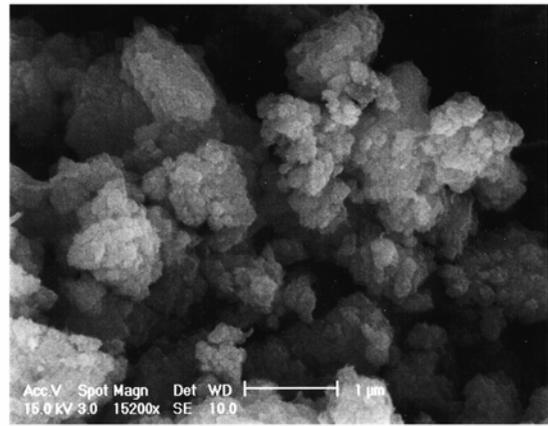


Fig. 3

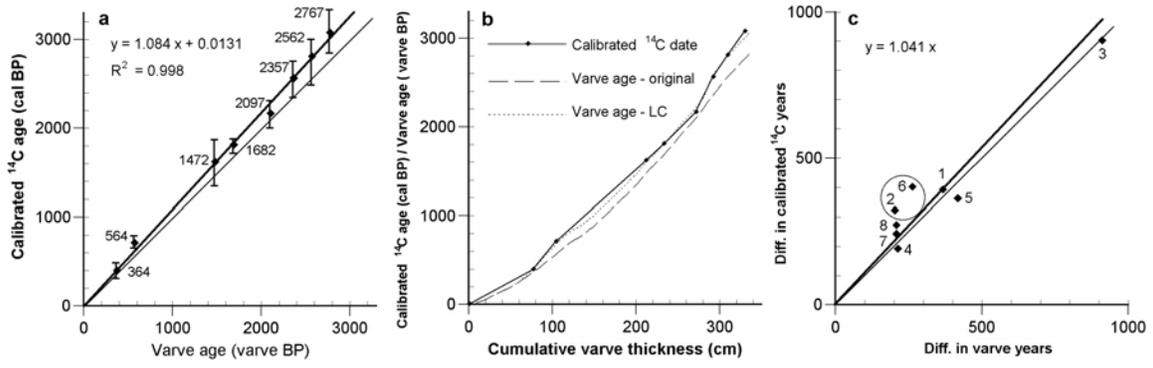


Fig. 4

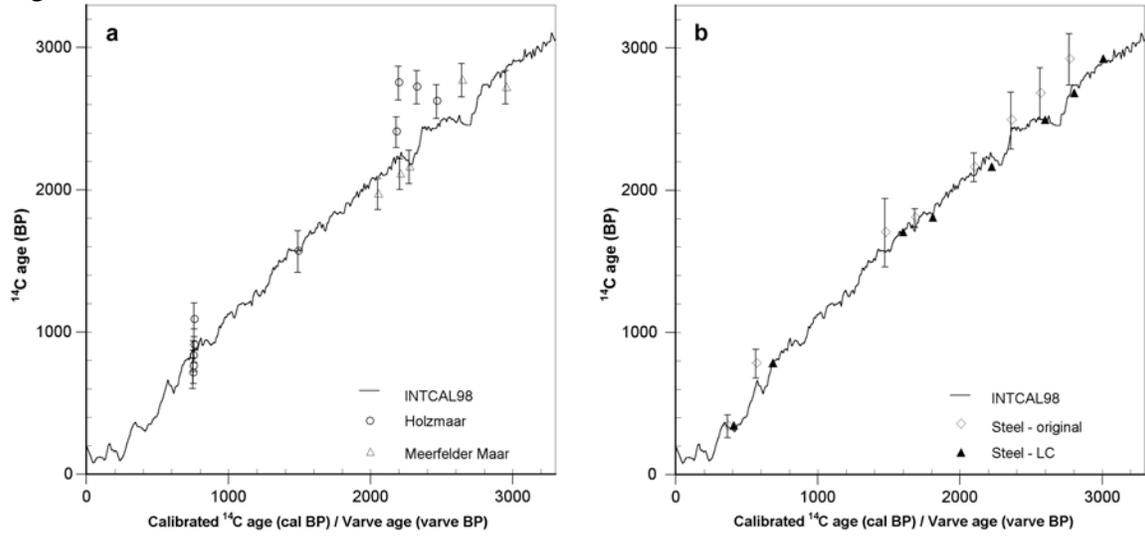


Fig. 5

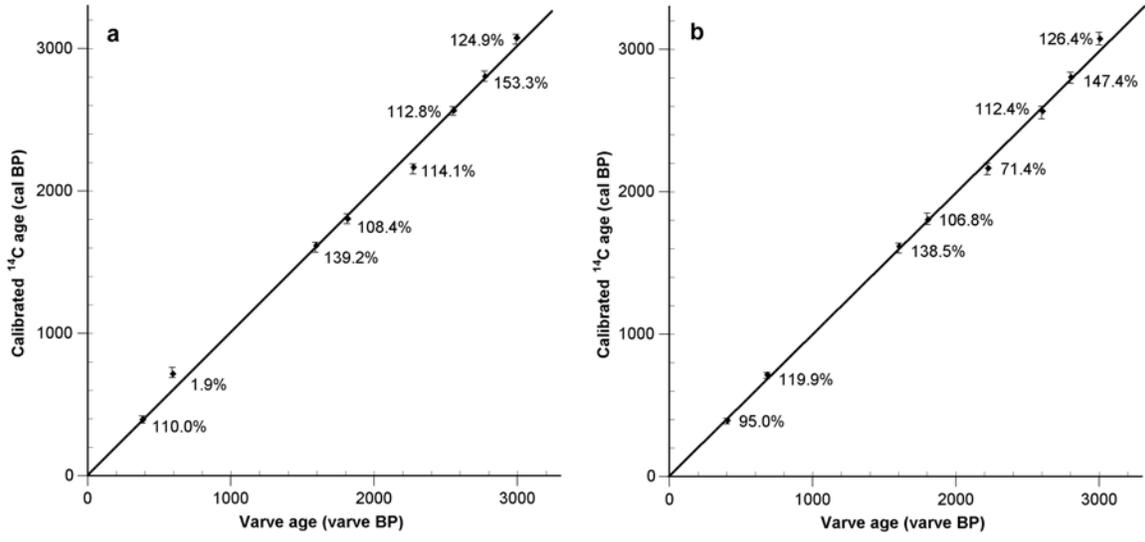


Fig. 6

