A long-term decrease in the persistence of soil carbon caused by ancient Maya land use

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Introductory Paragraph

The long-term effects of deforestation on tropical forest soil carbon reservoirs are poorly understood, but they are important for estimating the future consequences of human land use on the global carbon cycle. The Maya Lowlands of Mexico and Guatemala provide a unique opportunity to assess this question, given the widespread deforestation by the ancient Maya that began ~4000 years ago. Here we present past changes in the mean soil transit time of plant waxes (MTT_{wax}), determined by comparing the radiocarbon ages of plant waxes and plant macrofossils, in sediment cores collected from three lakes in the Maya Lowlands. Catchment-scale analyses of modern soils and lake sediments indicate that MTT_{wax} reflects the age of recalcitrant carbon in deep soils. All three sediment cores showed a decrease in $MTT_{wax,}$ ranging from 600 to 2200 years, from 3500 years ago to present. This decrease in MTT_{wax}, indicating reduced persistence of recalcitrant carbon in lake catchment soils, is associated with palynological evidence for ancient Maya deforestation. MTT_{wax} never recovered to pre-deforestation values, despite a subsequent decline in human population and reforestation, implying that current tropical deforestation will have long-lasting future effects on soil carbon sinks.

Main

Soil carbon is the largest terrestrial carbon reservoir (approximately 1500 Pg C)[1,2], and there is concern that it is being destabilized by climate and land use change[2-4]. Tropical forests contain about 30% of global soil carbon stocks (~470 Pg), of which about half is contained in subsoils below the organic-rich topsoil layer (typically >20-30 cm depth)[1,2,5]. Radiocarbon data indicate that tropical forest subsoils contain a sizeable proportion of slow-cycling carbon that persists for thousands of years[6-8]. Tropical forest soil carbon is at an especially high risk of destabilization because of widespread tropical deforestation over the past 50 years[3,9], but the impact of deforestation and land use on the ability of deep soil-carbon reservoirs to store carbon over long periods of time remains poorly constrained[10].

Ancient Maya land use provides an opportunity to evaluate the long-term effects of deforestation on carbon cycling in tropical forest environments. The low-elevation tropical forests of southeastern Mexico and northern Central America, i.e. the Maya Lowlands (Fig. 1), sustained large human populations between ca. 2500 and 1000 BP[11], and ancient Maya urbanization and agriculture led to widespread deforestation and soil erosion[12-15]. Furthermore, because Lowland Maya population declined substantially during the Terminal Classic period (~1250 to 1100 years BP), and following the Spanish conquest (~450 to 350 years BP)[11], the Maya Lowlands also experienced lengthy periods of reduced land use intensity. Previous studies in the Maya Lowlands applied palynological and sedimentological methods to infer the response of tropical forest ecosystems to land use change[12,13,15-17], but did not evaluate changes in soil carbon dynamics.

In this study we measured radiocarbon (^{14}C) in long-chain (C_{26}, C_{28}, C_{30}, C_{32}) *n*-alkanoic acids, which are derived from the leaf cuticular waxes of terrestrial vascular plants[18] (referred to here as plant waxes), in sediment cores from three lakes across the Maya Lowlands (Fig. 1). Stable isotope data confirm that the plant waxes in sediments from these lakes are derived from terrestrial plants[19,20] (see Methods), and plant wax production does not vary significantly among the major plant groups in these catchments, angiosperm trees and grasses[21]. We define the mean transit time of plant waxes in catchment soils (MTT_{wax}) as the mean age of plant waxes that are transported from soils to lake sediments at a given point in time[22]. We calculate MTT_{wax} as the difference between the age of plant waxes in sediments and the age of the sediment horizon in which they were buried, which is determined by ^{14}C analysis of plant macrofossils (see Methods). Previous studies of plant-wax ^{14}C values indicated that plant waxes are representative of slow-cycling soil carbon pools in terrestrial ecosystems[23-26], and MTT_{wax} values in sediment cores thus serve as an indicator of changes in the persistence of soil carbon across a catchment through time[27-29].

Plant-wax ^{14}C in modern catchment soils and lake sediments

We compared ^{14}C measurements of plant waxes and bulk soil organic carbon (SOC) in soils from the catchment of Lake Chichancanab to inform our interpretation of changes in MTT_{wax} in the sediment cores. Differences in $\Delta^{14}C$ between plant waxes and bulk SOC range between 9 to 51 ‰, which in subsoils (≥ 20 cm depth) corresponds to ^{14}C age differences between 70 to 440 years, respectively (Fig. 2). In topsoils, age differences are not easily determined because of the input of bomb-derived ^{14}C since 1950. Plant waxes are not consistently depleted in ^{14}C relative to bulk SOC, despite typically being among the oldest and most recalcitrant fractions of soil carbon[26]. Soils in which bulk SOC is older than plant waxes may contain other refractory carbon fractions, such as black carbon or petrogenic carbon[30]. Both plant waxes and SOC in subsoil horizons have negative $\Delta^{14}C$ values, indicating these carbon fractions are, on average, hundreds of years old. Plant waxes in particular exhibit progressively lower $\Delta^{14}C$ in deeper soils (Fig. 2). Bulk SOC $\Delta^{14}C$ exhibits a less consistent pattern with soil depth, which probably reflects local-scale heterogeneity caused by processes that include the input of root-derived carbon, preferential carbon flow pathways, and bioturbation[30]. Plant waxes have been found to be concentrated in mineral-bound SOC, which may account for the more consistent increases in age with soil depth[26].

Plant waxes in Lake Chichancanab surface sediments are ^{14}C-depleted relative to plant waxes in all catchment soil samples (Fig. 2), implying that plant waxes in the sediment are largely derived from deep soils, below 50 cm[20]. This may be a consequence of subsurface transport of organic matter in this karst watersheds[31], or to selective preservation of mineral-bound plant waxes in lake sediments[25]. These are currently the only data that enable comparison of plant wax ^{14}C in soils and sediments from the same catchment[20]. Although additional datasets would help to constrain the processes that control MTT_{wax} inferred from sediment cores, these results corroborate other studies that indicate plant waxes are representative of mineral-associated subsoil carbon[23,24,26], and that plant wax ages in sediments reflect the persistence of subsoil carbon in the surrounding catchment[25,32].

A long-term regional reduction in plant wax transit time

Records of MTT_{wax} from the three lake cores reveal broadly consistent patterns of change through time (Fig. 3A). Over the past 3500 years, a substantial decrease in MTT_{wax} was recorded in all three sediment cores, with the net change in mean transit time values (i.e., the difference between the highest and lowest value) ranging from 836±300 years at Lake Chichancanab to 2290±330 years at Lake Itzan.

The temporal pattern of the long-term decrease in MTT_{wax} differed among the three lakes. At Lake Salpeten there were three distinct periods of declining MTT_{wax}: during the Preclassic period (3400 to 2400 years BP), during the Classic and Postclassic periods (1700 to 500 years BP), and during the past 150 years. At Salpeten there were also intervals of increasing MTT_{wax} during the transition from the Preclassic to Classic periods (2400 to 1700 years BP), and following the Postclassic period (500 to 150 years BP). At Laguna Itzan, MTT_{wax} also declined sharply during the Preclassic period (3300 to 1900 years BP), but then increased gradually through the Classic and Postclassic periods (1900 to 540 years BP), before declining again over the past 150 years. The record at Lake Chichancanab is shorter, but indicates decreasing MTT_{wax} from the late Preclassic through the Postclassic period (2200 to 650 years BP), followed by increasing MTT_{wax} to the present.

There is a clear temporal association between deforestation, as indicated by regional pollen records (Figure 3E), and declining MTT_{wax} in the three lake catchments. The interval of pronounced decreasing MTT_{wax} during the Preclassic period at Salpeten and Itzan corresponds to the initial deforestation of the Maya Lowlands. The end of the Preclassic period was marked by a temporary reversal of deforestation (Fig. 3E) and possibly decreasing regional population[11,33], coinciding with stabilizing or increasing MTT_{wax} at Itzan and Salpeten. The Classic Period then brought renewed deforestation (Fig. 3E) and peak human population densities[11,33], coinciding with decreased MTT_{wax} at Salpeten and Chichancanab. Notably MTT_{wax} at Itzan was stable through the Classic period, possibly a consequence of reduced land use pressures or implementation of soil conservation practices within the catchment.

The transition from the Classic to the Postclassic period brought lower population density and reforestation, processes that were enhanced by the Spanish Conquest that began ca. 500 years BP[11,34]. At Salpeten and Chichancanab, MTT_{wax} only began to increase again following the Spanish Conquest. Finally, decreasing MTT_{wax} at Lake Salpeten and Laguna Itzan over the past 150 years coincided with the rapid, extensive 20th-century deforestation of these catchments and throughout northern Guatemala[35]. The region surrounding Lake Chichancanab did not experience such recent deforestation, likely explaining the absence of a recent decrease in MTT_{wax} there.

We also analyzed two deeper mid-Holocene horizons from a sediment core collected at Lake Salpeten[12] (Fig. 3B), which yielded MTT_{wax} estimates of 1500±225 and

1760±210 years. Although they do not provide a continuous record of MTT_{wax} change prior to anthropogenic deforestation, these mid-Holocene MTT_{wax} values indicate that the high values observed 3500 years ago at Salpeten and Itzan were not anomalous, and that there was not a continuous, long-term decrease in MTT_{wax} through the Holocene. This suggests that high MTT_{wax} values observed prior to major anthropogenic deforestation at ~3500 years BP (Fig. 3A) represented steady-state conditions that had persisted for thousands of years.

An alternative hypothesis is that late Holocene hydroclimate changes caused the observed changes in MTT_{wax}, as previous work indicate that changes in precipitation can alter MTT_{wax} in tropical forests[28]. There is not, however, a consistent relationship between paleohydrological records from the Maya Lowlands and MTT_{wax} (Fig. 3F). For example, regional paleoclimate records indicate a trend towards wetter conditions between 3000 and 2000 years BP[36], a time when we observed a pronounced decrease in MTT_{wax} at Salpeten and Itzan. In contrast, the regional climate became drier between 2000 and 1000 years BP, while MTT_{wax} decreased at Salpeten and Chichancanab. This inconsistent relationship implies that hydrological change was a secondary influence on MTT_{wax} in the Maya Lowlands during the late Holocene.

Mechanisms relating deforestation and plant wax transit time

There are multiple mechanisms by which ancient Maya deforestation could have caused the observed reduction in MTT_{wax}. Persistence of SOC is controlled largely by protection from microbial biodegradation, either through sorption to mineral surfaces or occlusion in soil aggregates[10,30,37]. Mineral sorption of SOC onto Al- and Fe-bearing minerals is typically favored in acidic soils, whereas Ca-mediated aggregate formation is the dominant mode of soil carbon preservation in basic soils[37]. Soils in the karstic Maya lowlands form through the progressive weathering of the carbonate bedrock, and this process entails gradual concentration of Al and Fe supplied by a combination of minor autochthonous minerals and allocthonous inputs from dust and volcanic ash[14,38]. Maya deforestation disrupted bedrock weathering and greatly enhanced soil erosion[12,14]. Preserved soils that predate deforestation show higher concentrations of Al and Fe and lower pH (as low as 5.8), compared with younger soils formed following Maya deforestation (with pH as high as 8.4)[14]. This change in regional soil chemistry, associated with deforestation, probably reduced the capacity for mineral sorption in soils, and as pH rose to values near neutral, rates of microbial decomposition may have increased[37]. *N*-alkanoic acids are concentrated in mineral-bound organic matter[26], and therefore reduced mineral sorption in the soils of the Maya lowlands soils would have been particularly effective in reducing MTT_{wax}.

Disruption of soil aggregates is also associated with deforestation and farming, through a variety of mechanisms that include tillage, increased soil disruption through tree removal and crop harvesting, and interference with of root and fungal networks that promote aggregation[39,40]. Deforestation in the Maya Lowlands probably led to both reduced mineral sorption and disruption of soil aggregates, both of which would have led to a reduced persistence of soil carbon and lower MTT_{wax}, though the relative importance of these two mechanisms is uncertain. A potential modern analog for the changes in soil carbon storage in the Maya Lowlands is recent deforestation in the karst terrain of southwest China, which has been associated with increasing soil pH[41], the loss of carbon from soil macroaggregates[39], and a pronounced loss of carbon from deep soils, below 50 cm[42].

The transport of eroded soil and its effects on MTT_{wax} also require consideration, as there is sedimentological evidence of enhanced soil erosion related to ancient Maya deforestation during the Preclassic period, in both the Salpeten[12] and Itzan catchments (Fig. 3D), although not at Chichancanab[43]. As soil erosion progressed, it is possible that surface transport of deep subsoil carbon, which contain old plant waxes, would lead to increased MTT_{wax} values, as was inferred in previous studies[27,28]. However, this is not consistent with our results, as increased soil erosion at Salpeten and Itzan between 3500 and 2000 years BP (Fig. 3D) coincided with decreasing, rather than increasing MTT_{wax} (Fig. 3A). Furthermore, as soil erosion increased during the Preclassic there was a net decrease in the mass accumulation rate of plant waxes (MAR_{wax}; Fig. 3C; see Methods) at Salpeten, inconsistent with an increasing erosive input of plant waxes to the lake sediments. At Itzan, MAR_{wax} first increased along with soil erosion between 3300 and 2600 years BP, but then MAR_{wax} decreased to its lowest levels, whereas soil erosion remained at high levels between 2600 and 1900 years BP. In general, there is an inconsistent relationship between MAR_{wax} and MTT_{wax} in the three lake cores, suggesting that erosive transport of plant waxes was probably not a primary control on the long-term decrease in MTT_{wax}.

Soil erosion can in some cases enhance the persistence of soil carbon by burying older soils under freshly eroded material[44]. However, the depositional burial of older soils, which could have potentially isolated plant waxes in older carbon reservoirs and prevented their transport to lake sediments, is unlikely to have caused the observed decrease in MTT_{wax}. The Salpeten and Itzan catchments are small and possess relatively steep slopes[20], and do not contain significant areas for eroded soils to accumulate aside from the lakes[12]. Furthermore, our comparison of modern sediments and soils at Lake Chichancanab indicated that while MTT_{wax} in modern sediments is relatively low compared to peak values in deeper sediments, plant wax ^{14}C ages in modern sediments are older than those found in any soil samples (Figure 2). This suggests there is not a reservoir of old plant waxes in soils that is isolated from the flux transported to the lake sediments.

Implications for past and future soil carbon dynamics

Estimates of pre-industrial land use effects on soil carbon storage are poorly constrained[45], in large part because existing soil carbon models do not include long-term responses of soil carbon pools to environmental change[10]. Comparison of the minimum and maximum MTT_{wax} values calculated for each of our lake sediment cores suggests that Maya deforestation reduced the transit time of plant waxes, and likely other mineral-associated subsoil carbon, by between 70% (Chichancanab) to 90% (Salpeten and Itzan) relative to pre-deforestation values. This implies that Maya land use strongly reduced the ability of regional soils to act as long-term carbon sinks.

Comparison of our data with sediment core plant-wax ^{14}C data from Saanich Inlet, British Columbia[29], Lake Soppensee in Switzerland[27], and the Congo River Fan[28] (Fig. 4) indicates substantial geographic and temporal variability in MTT_{wax} over the past ~6000 years. Detailed comparison of these records is difficult given the large differences in area, climate, geology, and geomorphology of the sedimentary catchments. There is clearly substantial temporal variability in MTT_{wax} in all of these records, however, with values varying by more than 2000 years. This geographically widespread temporal variability in MTT_{wax} suggests that the persistence of slow-cycling soil carbon has been highly dynamic on millennial time-scales during the Holocene, particularly during the past ~6000 years. This dynamism in soil carbon reservoirs should be validated in other environments and could be important for models of Holocene carbon cycling[45,46]. Data from the Maya Lowlands and Switzerland, in particular, indicate that pre-industrial human land use profoundly changed soil carbon reservoirs, which may represent a substantial proportion of the pre-industrial anthropogenic impact on the global carbon cycle[45,47].

The protracted decrease in MTT_{wax} in the Maya Lowlands over the past 3500 years suggests that extensive modern tropical deforestation will have profound long-term effects on future carbon storage in tropical forest soils. Reductions in MTT_{wax} in the Maya Lowlands over the past 150 years are small compared with decreases caused by ancient Maya land use (Figure 3A). The first episode of declining MTT_{wax} between ca. 3500 and 2500 years BP, which coincided with the initial deforestation of the Maya Lowlands, may be a better analogue for the recent, ongoing deforestation of primary forest in many tropical regions. In general, our data indicate that MTT_{wax} decreased progressively over hundreds to thousands of years (Fig. 3A), suggesting that tropical forest soil carbon reservoirs, at least in subsoils, do not reach equilibrium within a few decades following deforestation[9]. Instead, our results suggest that periods of sustained deforestation and agriculture lead to long-term decreases in the persistence of subsoil carbon over centuries to millenia.

MTT_{wax} in the Maya Lowlands has never recovered to the high values that preceded initial deforestation, despite lengthy periods of reforestation and soil stability[12,16] (Fig. 3). This suggests that prolonged periods of time are necessary to accumulate stable, slow-cycling carbon pools in reforested tropical soils, particularly if past land use changed to soil chemistry in ways that diminish mineral sorption or soil aggregation[14,37,39]. These results suggest that the effects of primary tropical forest removal on soil carbon reservoirs are not offset by growth of secondary forest, at least in terms of long-term carbon storage in subsoils. This, in turn, implies that to maximize long-term soil carbon storage, it is more effective to conserve primary forests than to reforest previously deforested areas[48].

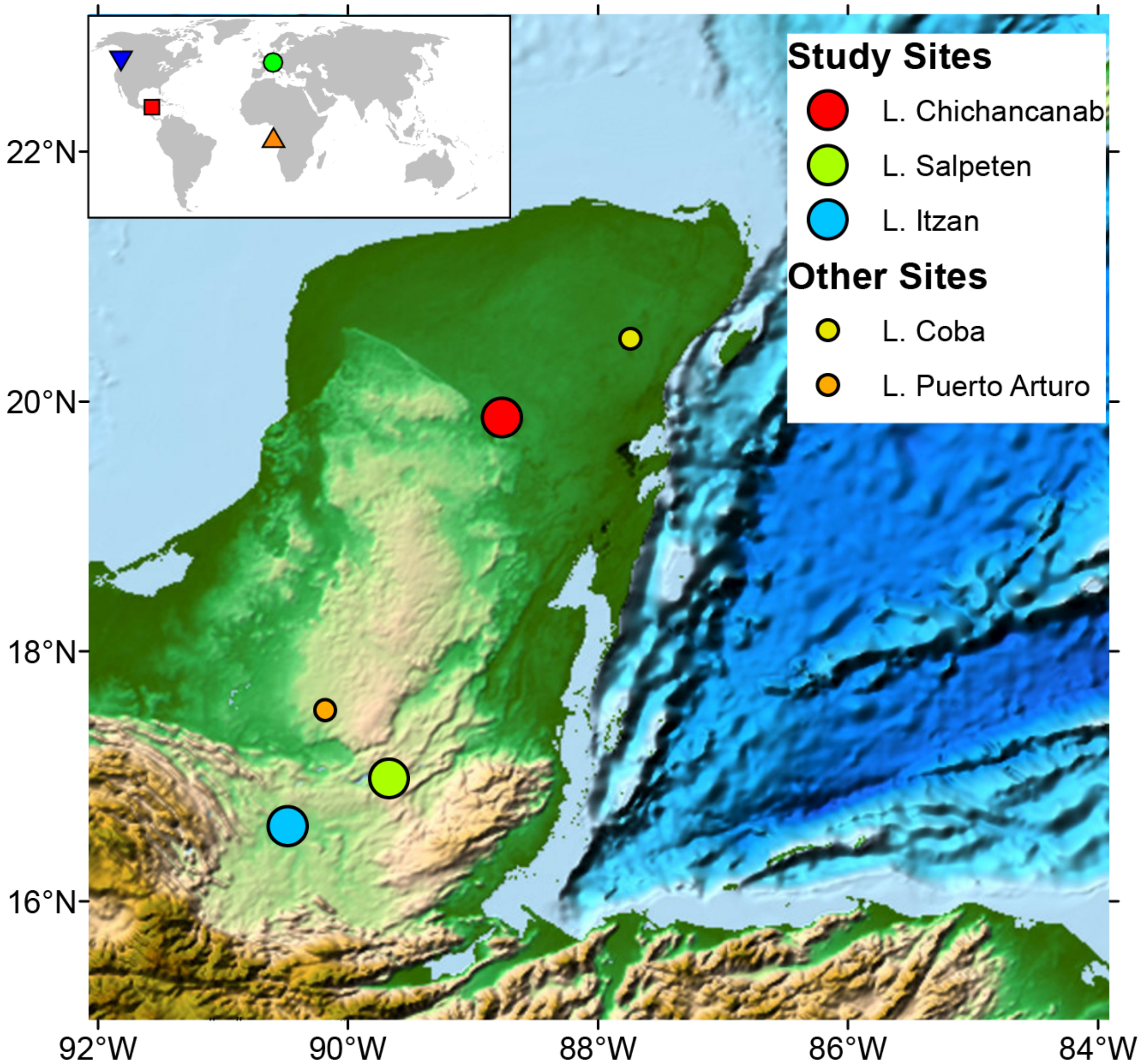


Figure 1 Relief map of the Maya Lowlands indicating the location of the studied lakes. Additional lakes studied for palynological records are also shown. The inset map shows the location of the Maya Lowlands (red square) relative to Saanich Inlet, Canada (blue triangle), Lake Soppensee, Switzerland (green circle), and the Congo River Fan (orange triangle) (see Fig. 4).

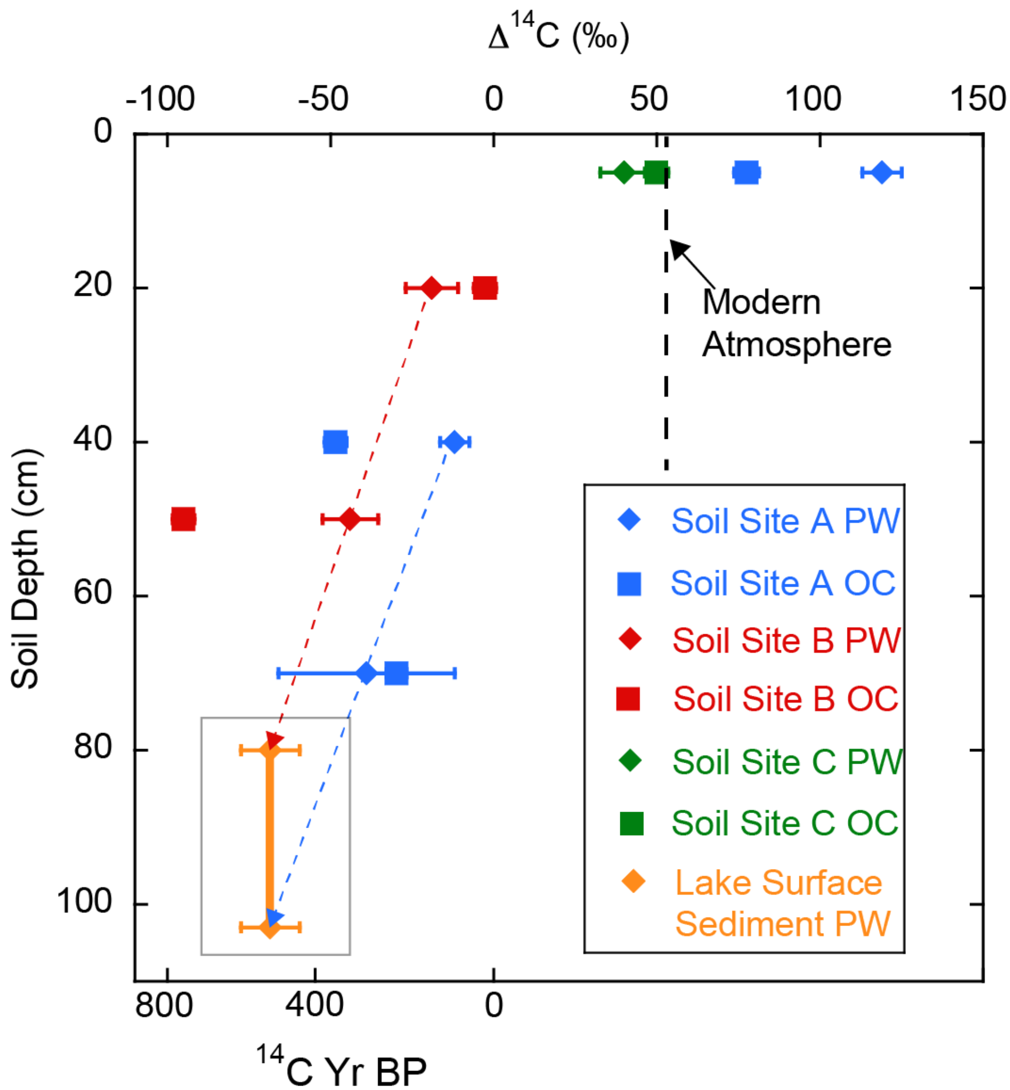
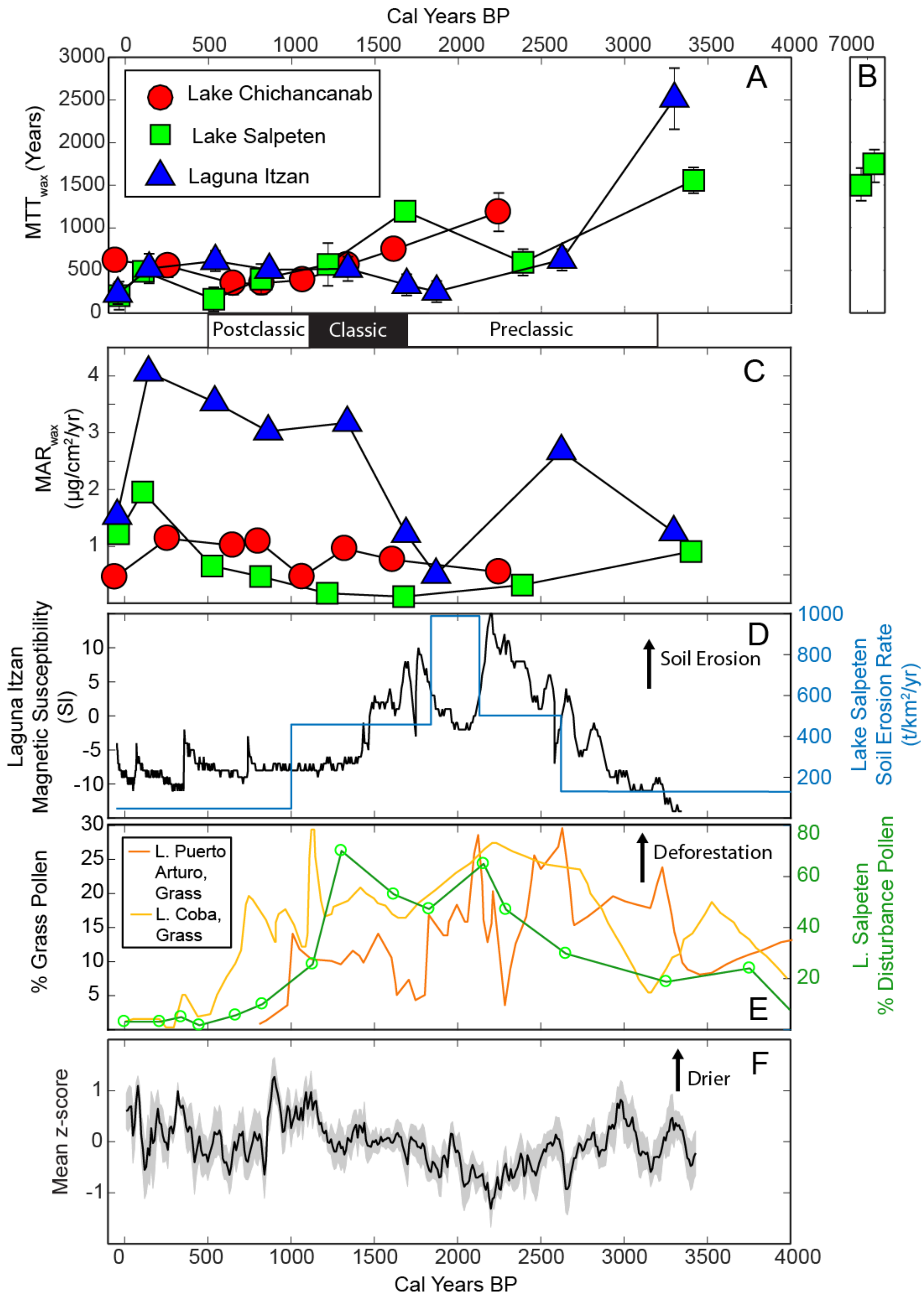


Figure 2 Plant-wax (PW) and bulk soil organic carbon (SOC) radiocarbon results from soils and surface sediments in the Lake Chichancanab catchment. Both plant waxes and bulk SOC generally exhibit greater age with soil depth, a trend that is more consistent for plant waxes. Plant-wax $\Delta^{14}C$ values from the lake surface sediments are included on the same plot to indicate the depth in the soil profile at which soil plant waxes would have equivalent $\Delta^{14}C$ values, assuming a linear decrease in plant-wax $\Delta^{14}C$ with subsoil depth at a given location.



Cal Years BP
A
B
Lake Chichancanab
Lake Salpeten
Laguna Itzan
MTT$_{wax}$ (Years)
Postclassic
Classic
Preclassic
C
MAR$_{wax}$ (μg/cm²/yr)
D
Soil Erosion
Laguna Itzan Magnetic Susceptibility (SI)
Lake Salpeten Soil Erosion Rate (t/km²/yr)
E
Deforestation
L. Puerto Arturo, Grass
L. Coba, Grass
% Grass Pollen
L. Salpeten % Disturbance Pollen
F
Drier
Mean z-score
Cal Years BP

275

276

Figure 3 Records of MTT_{wax} **(A)** and comparison with environmental change in the Maya Lowlands. (**B**) shows mid-Holocene MTT_{wax} from Lake Salpeten. MTT_{wax} error bars indicate 95% confidence intervals. Approximate dates of Maya archaeological periods are indicated below (**A**). Plant-wax mass accumulation rates (MAR_{wax}) are shown in (**C**). Lake Salpeten soil erosion rates (**D**) are derived from sediment accumulation measurements[12]; Laguna Itzan magnetic susceptibility also reflects soil erosion. Pollen records (**E**) from three lake cores reflect regional trends in deforestation[13,15,17]. The composite paleohydrological record for the Maya Lowlands **(F)** was calculated from z-scores for eight paleolimnological and speleothem paleoclimate studies[36].

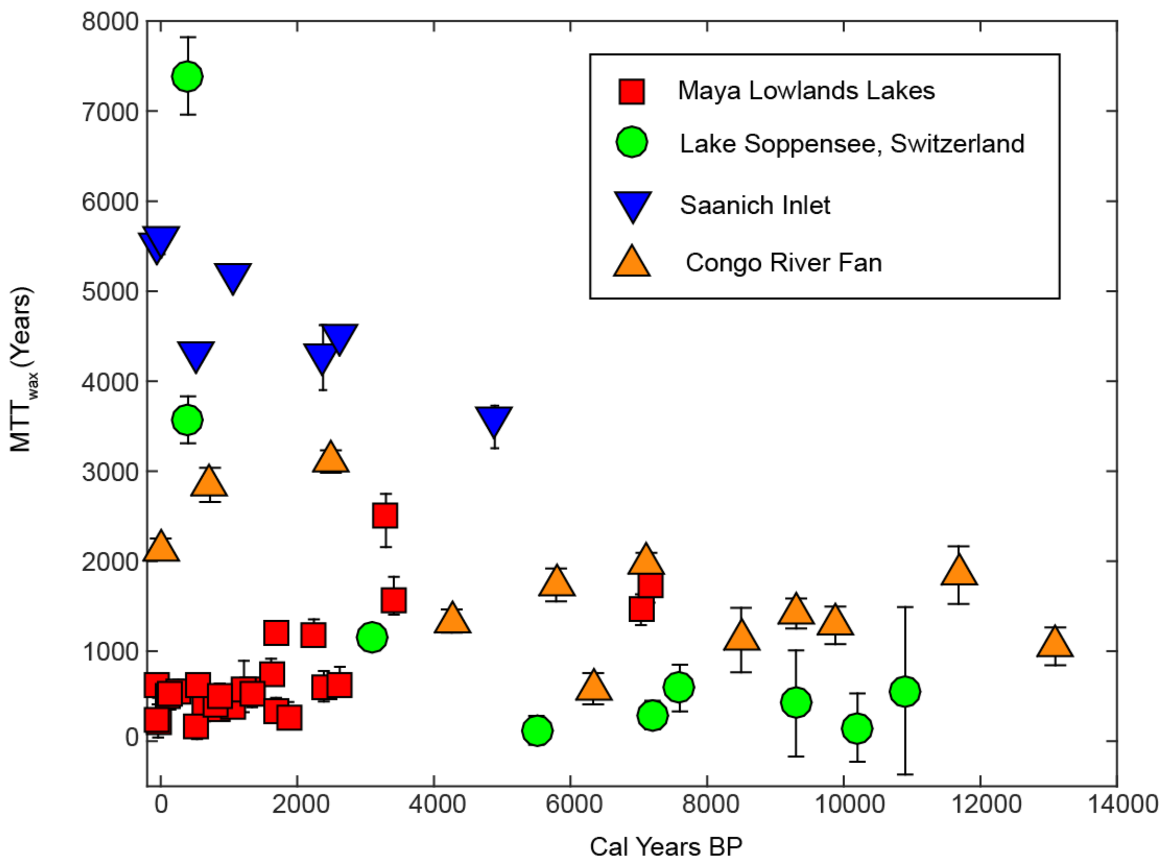


Figure 4 Comparison of MTT_{wax} data from the Maya Lowlands with three other globally distributed Holocene-age records[27-29]. These data suggest widespread changes in the transit time of plant waxes in soils, particularly during the past 6000 years, although they likely differ with respect to the causes of these changes.

Methods

Study Sites

Lakes Chichancanab, Salpeten, and Itzan (Fig. 1) are located in representative low-elevation tropical environments inhabited by the ancient Maya. All three are relatively small, have limited catchments (137 to 1.5 km^2)[20] and are located in the karst terrain that characterizes most of the Maya Lowlands[49]. Lakes Salpeten and Itzan contained substantial ancient Maya populations within their catchments[33,50]. For example, the human population of the Lake Salpeten catchment is estimated to have peaked at around 4000 people during the Late Classic period, ca. 1200 BP[33]. Lake Chichancanab was farther from major Maya population centers and its catchment was probably less densely populated[43], although analysis of pollen in a sediment core from the basin indicates ancient maize agriculture occurred in the catchment[51]. Modern land use also differs among the lake catchments. There has been extensive recent forest clearance in the Lake Salpeten and Laguna Itzan catchments, but relatively less modern deforestation in the Lake Chichancanab catchment. The lakes vary in terms of precipitation, with the greatest mean annual rainfall at southernmost Laguna Itzan (2098 mm) and the least at northernmost Lake Chichancanab (1161 mm)[20,52]. This difference in rainfall is reflected in the natural vegetation, with denser, higher-stature forest and a greater proportion of evergreen trees in the remaining natural vegetation around Laguna Itzan and Lake Salpeten, and lower-stature, more open and seasonally deciduous forest at Lake Chichancanab[53]. Soils surrounding Lake Chichancanab are primarily rendzic leptosols, whereas soils in the Laguna Itzan and Lake Salpeten catchments are primarily cambisols[38,54]. Soil depths are not thoroughly mapped in the Maya Lowlands. In the northern Yucatan Peninsula around Lake Chichancanab soils are typically shallow (0.5 to 1 m), but deeper soil profiles probably occur at the wetter sites in Guatemala[14,38].

Sediment and soil sampling

Sediment coring at Lake Chichancanab and Lake Salpeten was described previously[43,55,56]. Middle Holocene sediments from Lake Salpeten were sampled from a 14 m sediment core collected in 1980[15], which was age-depth correlated with the cores collected in 1999 from which the other samples were taken[12].

Two overlapping cores, totaling 5.7 m of sediment, were collected in 1997 from Laguna Itzan, at a water depth of 10 m, near the western shore of the lake. The upper 2.2 m of sediment is dominated by silt-sized carbonate mud. Between 2.3 and 5.5 m depth the sediment alternates between silt-size carbonate mud and calcite-rich clay. Magnetic susceptibility of the Laguna Itzan cores was measured using a Geotek Multi-Sensor Core Logger at the University of Minnesota Limnological Research Center in 1997 (Supplemental Table S6). Magnetic susceptibility reflects soil erosion, as it is controlled by the input of magnetic, iron-bearing minerals from soils into lake sediments.

In all sediment cores, samples for plant-wax radiocarbon analysis were collected from stratigraphic horizons as close as possible (< 2 cm), adjacent to dated terrigenous macrofossils. In the Lake Salpeten and Chichancanab cores, however, we also analyzed plant-wax ^{14}C in horizons for which there were no adjacent terrigenous macrofossils to distribute plant-wax ^{14}C ages throughout the sediment cores.

In December 2012, soil samples were collected from sites around Lake Chichancanab[20]. Sites A and B are located in forested uplands approximately 15 and 24 m above lake level, respectively. Site C is near the lakeshore, < 1 meter above lake level, and is inundated during periods of high water. At each site, a pit was dug and samples were collected from the pit wall, with care taken to avoid contamination from overlying horizons. Subsoil samples (<20 cm depth) from site C did not contain sufficient quantities of long-chain fatty acids for $\Delta^{14}C_{wax}$ analysis, and were not studied further.

Compound-specific radiocarbon analyses

Sediment and soil samples were freeze-dried and solvent-extracted (ASE3000, Dionex). Total lipid extracts were then saponified, and the acidic fraction was transesterified and purified via silica gel chromatography. Purified *n*-alkanoic acids, analyzed as fatty acid methyl esters (FAMEs), were quantified relative to an external standard using a gas chromatograph coupled to a flame ionization device. Long-chain *n*-alkanoic acid methyl esters were isolated using a preparative capillary gas chromatography system[57]. Individual *n*-alkanoic acid homologs were not sufficiently abundant for radiocarbon analyses, so we measured the combined C_{26}, C_{28}, C_{30}, and C_{32} homologs. Isolated *n*-alkanoic acid samples were combusted to CO_2, graphitized, and analyzed for radiocarbon at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) Facility, Woods Hole, MA, and USA.

All compound-specific radiocarbon data were corrected to account for the estimated procedural blank associated with sample extraction and purification[58], and they also were corrected for the methyl carbon added during transesterification, which contains no measurable ^{14}C. The procedural blank determined for the WHOI Marine Chemistry and Geochemistry PCGC system (which was used to isolate the Lake Chichancanab samples) was 1.8±0.9 μg of C with an Fm of 0.44±10[58], whereas the procedural blank for the NOSAMS PCGC system (used to isolate all other samples) was

1.4±1.2 μg of C with an Fm of 0.64±0.2[20]. Compound-specific radiocarbon data for Lakes Salpeten and Chichancanab were previously published[19,20], with the exception of mid-Holocene samples from Lake Salpeten (Table S1). Compound specific radiocarbon data from Laguna Itzan are presented in Table S1.

In addition to terrestrial vascular plants, *n*-alkanoic acids can be derived from aquatic plants[59,60], which could potentially influence the interpretations of changes in MTT_{wax} presented here. Stable isotope analyses of *n*-alkanoic acids in aquatic plants from the Maya Lowlands, however, indicate that they are significantly depleted in δD and $\delta^{13}C$ relative to plant waxes found in both regional soils in the northern Yucatan Peninsula and in sediments from Lake Chichancanab and Salpeten[19,20]. This implies that aquatic plants are not an important source of plant waxes in sediments from these lakes and do not have a significant effect on plant-wax ^{14}C values. At Laguna Itzan we observed similarly enriched *n*-alkanoic acid $\delta^{13}C$ values in the deeper sediment samples, but more depleted $\delta^{13}C$ values that overlap with the range of aquatic plants in the four uppermost sediment samples, from 930 years BP to present (Table S1). However, these samples postdate the major decrease in MTT_{wax} in this sediment core, and therefore possible input from aquatic plants in these sediment samples would not significantly alter our interpretations. Furthermore, these values are also consistent with plant-wax $\delta^{13}C$ values observed in soils from moist tropical forest soils in southern Guatemala[59] whose climates are similar to that at Laguna Itzan, as well as in plants in other low-elevation tropical forest environments[61]. Thus the low $\delta^{13}C$ values also are consistent with a dominantly terrestrial plant source for long-chain *n*-alkanoic acids.

Terrigenous macrofossil radiocarbon analyses

For Lakes Chichancanab and Salpeten we compared plant-wax radiocarbon ages with previously published terrigenous macrofossil radiocarbon ages[43,55,62,63]. To establish a chronology for sediment deposition at Laguna Itzan we analyzed the radiocarbon content of 11 terrigenous macrofossil samples, either leaf or wood fragments, distributed throughout the upper six meters of the sediment core (Supplemental Table S2). These samples were pretreated using a standard acid-base-acid protocol to remove carbonate minerals and humic acids[64], and they were analyzed for radiocarbon content at either the Lawrence Livermore National Laboratory or DirectAMS accelerator mass spectrometry laboratories.

Bulk soil organic carbon isotope analyses

Soil samples were sieved (2 mm) and freeze-dried, and visible plant matter was removed. Samples were then homogenized and soaked in 0.5 M hydrochloric acid for 24 hours to remove carbonate[65], followed by repeated rinses in Milli-Q water to remove acid salts. The pretreated samples were analyzed for radiocarbon content at DirectAMS (Supplemental Table S3). In addition to radiocarbon analyses, we analyzed bulk soil $\delta^{13}C$ values using a Costech Elemental Analyzer coupled to a Thermo DeltaPlus Advantage Isotope Ratio Mass Spectrometer at the Yale University Earth System Center for Stable Isotope Studies (Supplemental Table S3).

Sediment core age-depth models

For each core, we constructed two age-depth models, one for the age of sediment deposition and one for the mean age of plant-wax synthesis (Fig. S1). Sediment deposition age models were based on radiocarbon ages of terrigenous plant macrofossils, which is a common practice in tropical lakes in carbonate terrain, because macrofossils are typically transported rapidly to sediments and their ^{14}C dates are not affected by hard-water lake error [55,62]. Plant-wax age models were based on their compound-specific radiocarbon ages. For all age models, we applied the Classical Age-depth Modeling (CLAM 2.1) software in R[66]. All radiocarbon ages were calibrated using the IntCal13 calibration curve[67]. 95% confidence intervals for age-depth models were calculated by analyzing the distribution of 1000 randomly generated age models[66]. The 'best' age model was determined by calculating the mean age of all model iterations at each depth in the core.

For the Lake Chichancanab and Lake Salpeten sediment cores, we applied 2nd and 4th order polynomial age-depth models, respectively, as applied in the original studies of these cores[43,55] (Fig. S1 A,B). The best fit to the Laguna Itzan terrigenous macrofossil radiocarbon ages was provided by a 4th order polynomial age-depth model (Fig. S1 C). In all cores, core-top sediments were assumed to have been deposited in the year of core collection. For the middle Holocene samples from Lake Salpeten, sediment deposition ages were derived from a previously published linear interpolation age model[12]. To develop plant-wax age-depth models at Lakes Chichancanab and Laguna Itzan we fit a smoothing spline with a smoothing factor of 0.3 to the $^{14}C_{wax}$ ages (Fig. S1 A,C). At Lake Salpeten the best-fit age-depth model for plant waxes was provided by linear interpolation between dated horizons (Fig. S1 B).

The mean transit time of plant waxes in catchment soils (MTT_{wax}) was calculated as the age difference, for each depth in the core, between the plant-wax and sediment age-depth models. We applied the difference in the calibrated ages of the age-depth models because we do not have macrofossil ^{14}C ages for all core depths where we measured plant wax ^{14}C ages. MTT_{wax} represents the mean age of plant waxes at the time of sediment deposition. This calculation assumes that transit times between plants and catchment soils and from soils to lake sediments is rapid. This assumption is reasonable given that plant waxes are derived predominantly from leaves[68], which turnover within a few years[69], as well as the small catchment sizes, shallow lake water depths (10 to 32 m) and absence of apparent intermediate reservoirs[20]. MTT_{wax} confidence intervals were estimated by propagating the 95% confidence intervals for the plant-wax and sediment age-depth models (Fig. S1). MTT_{wax} values and the sediment and plant-wax ages used to calculate them are shown in Supplemental Table S4.

Calculation of plant-wax mass accumulation rates

To calculate MAR_{wax} we used the general equation:

$$MAR_{wax} = c_{wax} \times SAR \times DBD \quad (1)$$

where c_{wax} is the concentration of plant waxes in lake sediments (µg C_{26}, C_{28}, C_{30}, and C_{32} alkanoic acids per g dry sediment, measured using gas chromatography with a flame ionization detector in comparison with an external FAME standard), SAR is the sediment accumulation rate determined from the terrigenous macrofossil age-depth models (Fig. S1), and DBD is the dry bulk density of core sediments (g dry sediment per cm^{-3} wet sediment). Values for all of these sediment variables are shown in Supplemental Table S5.

For the Lake Chichancanab core, wet bulk density (WBD; g wet/cm^3 of wet sediment) data were available[43], but we did not have data for sediment porosity or water content, and so we used the following equation to estimate dry bulk density (DBD):

$$DBD = WBD - (1.025 * 0.9) \qquad \text{(Equation 1)}$$

where 1.025 represents the density of saline lake waters and 0.9 is an estimate for the porosity of the organic-rich gyttja sediments, based on studies from other lakes[12,70]. An assumption of constant porosity is valid at this lake, where sediment lithology varies little[43].

For the Lake Salpeten core, for which WBD data were not available, we estimated DBD using the following formula[71]:

$$DBD = \frac{D(2.5I + 1.6C)}{D + (1 - D)(2.5I + 1.6C)} \qquad \text{(Eq. 2)}$$

where D is the proportion of dry mass in wet sediment, I is the inorganic proportion of dry material (density ~ 2.5 g/cm^3), and C is the organic proportion of dry material (density ~ 1.6 g/cm^3). Whereas 1.6 g/cm^3 was the density of organic matter used in the original application of this equation, organic matter can have lower densities. Nevertheless, even using a density of 1.25 g/cm^3 produced negligible changes in our calculated MAR_{wax} values. D, I, and C were determined based on water content analyses and weight loss on ignition at 550 °C using a sediment core from Lake Salpeten collected in 1980[72], and were correlated to sediment depths in the 1999 Lake Salpeten sediment cores analyzed in this study using a composite age model[12,55].

At Laguna Itzan we estimated DBD by sampling 2 cm^3 of wet sediment at 10-cm intervals throughout the sediment core, drying the sediment in a 70° C oven, and measuring the dry sediment mass. It was not possible to perform this procedure in the uppermost meter of core sediments because of long-term drying of stored sediment cores. For these horizons, we estimated dry bulk density by applying a 2nd order polynomial regression between WBD (measured using a Geotek multi-sensor core logger), and DBD estimates for lower core sediments described above (R^2 = 0.75).

Within-core variation in MAR_{wax} is dominantly controlled by c_{wax}, and is not strongly correlated with SAR or DBD (Supplementary Table S5). Therefore we consider the temporal trends within a particular record, which is our primary focus, to be robust. Given the different approaches to calculating sediment density for each core, differences in absolute MAR_{wax} values between the lake sediment cores should be interpreted cautiously.

Data Availability Statement

The data that support the findings of this paper are available either as tables in the supplementary information file or in tables published in the peer-reviewed articles cited in the Methods. All data are available in a spreadsheet format upon request from the corresponding author.

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