



The process of leaf-wax transport to tropical lake sediments and its significance for inferring past changes in climate and carbon cycling

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Compound-specific hydrogen (δD) and carbon ($\delta^{13}C$) isotope measurements of terrestrial plant leaf waxes in lake sediments can be used to infer past hydrological and ecological changes in terrestrial environments. Transport of leaf waxes from terrestrial ecosystems to sediments, however, is a complex process that remains poorly understood. This is illustrated, for instance, by compound-specific radiocarbon analyses of leaf waxes in some marine and lake sediments that indicate millennial-scale time lags between compound synthesis on land and deposition in sediments. Uncertainty regarding leaf-wax transport requires that compound-specific radiocarbon analyses be undertaken as part of sediment core leaf-wax studies to a) establish chronological control for leaf-wax stable isotope records, and b) examine temporal variability in the soil residence time of leaf waxes.

Here we present compound-specific δD , $\delta^{13}C$ and $\Delta^{14}C$ measurements of long-chain n-alkanoic acids (leaf waxes) from a late Holocene sediment core from Lake Chichancanab, Yucatan Peninsula, Mexico. This core was previously studied using mineralogy (gypsum) and stable isotopes ($\delta^{18}O$ shell carbonate) to infer past climate. Both climate proxies indicated drought conditions during the Terminal Classic Collapse of the Maya civilization, from ~ 1250 to 1000 years BP. A leaf-wax δD record constructed using the core chronology developed with ^{14}C dates on terrestrial macrofossils indicated major hydrologic variability, but was not coherent with the other climate proxy records. Leaf-wax $\Delta^{14}C$ ages from seven sediment core depths are 400-1200 years older than ages on co-occurring terrigenous macrofossils. Fitting the leaf-wax δD record to a "leaf-wax chronology" based on compound-specific radiocarbon ages provides much better agreement with the mineralogical and oxygen isotope data. In particular, the revised record indicates a large positive shift in leaf-wax δD ($+40\%$) during the Terminal Classic Collapse, suggesting pronounced drought at that time. Similarly, a leaf-wax $\delta^{13}C$ record fit to the "leaf-wax ^{14}C chronology" shows agreement with regional pollen records, with a -1.9% shift in leaf-wax $\delta^{13}C$ during the Terminal Classic, coincident with reforestation and a decrease in maize pollen.

Leaf-wax $\Delta^{14}C$ measurements provide information on the soil residence time of leaf waxes, as indicated by the offset between leaf-wax and terrigenous macrofossil radiocarbon ages. Leaf-wax residence times declined markedly from ~ 1200 years at 2500 BP to ~ 420 years at 1100 BP, stabilized at ~ 475 years from 1200 to 700 BP, and then increased to ~ 700 years at present. This record suggests that the residence time of soil refractory organic carbon decreased during a period of sustained land use by the Preclassic and Classic Maya, then stabilized and increased following societal collapse and reforestation. The correlation between leaf-wax residence times and societal change suggests that Maya land use practices had a major influence on soil organic carbon dynamics in the Yucatan Peninsula during the late Holocene.

Iterative box modeling experiments provide additional constraints for the transport of leaf-waxes to Lake Chichancanab sediments. Box model results indicate the fractional contribution and residence time of leaf-waxes transported through organic carbon pools with annual-, decadal- and millennial-scale residence times. These modeling efforts yield further insights into the age distribution of leaf waxes in sediment archives, the chronology of leaf-wax stable isotope records, and the significance of leaf-wax radiocarbon ages for understanding long-term trends in refractory organic carbon cycling.