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Latitudinal and temporal patterns in terrestrial photosynthetic discrimination assessed by the carbon isotopic composition of plant leaf wax aerosols

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The molecular and isotopic signatures of biogenic compounds in aerosols provide a unique approach for the study of the terrestrial biosphere and climate-biosphere interactions on large spatial scales. Here we use the carbon isotopic composition of ablated plant leaf waxes in aerosols to resolve spatial and temporal patterns in carbon isotopic discrimination (Δ) of terrestrial ecosystems. We sampled bulk aerosols (2-week integration period) at strategically located sites (northern Alaska, southern Alberta, central Maine, Bermuda, central Florida, Barbados, northeastern Amazon) that receive boundary layer air masses advected across distinctive ecosystem types. The wax aerosol-derived Δ (using concentration-weighted mean of C24-34 n-alcohols) ranges from a nearly pure C4 plant signal ($\sim$8‰ in North African air masses sampled at Barbados in late autumn to a nearly pure C3 plant signal (19-20‰ in North American air masses that sample emissions from temperate forest ecosystems in late spring. Seasonality in wax aerosol-derived Δ is high (6-8‰) in Florida, Bermuda and Barbados air masses, whereas seasonality is low (<2‰) in Alaska, Maine and northeastern Amazon air masses. Short-term variability in wax isotopic composition that is associated with shifting air mass trajectories demonstrates the large-regional scale of the wax aerosol "footprint". The annually-integrated wax aerosol-derived Δ compares well with model estimates of the latitudinal distribution of Δ. An influence of biomass burning is also demonstrated by the seasonal rise to a nearly pure C4 plant signal observed at Barbados each Oct-Dec which coincides with the annual harvest/burning of C4 cereal crops in subSahara Africa. In contrast, wax aerosol concentration and isotopic composition at Florida, Bermuda, and Barbados do not vary with dust transport to these locations, indicating that Saharan dust is not the primary vector for wax atmospheric transport.