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Large spatial-scale and temporal variability in carbon isotopic fractionation of atmospheric CO_2 by the terrestrial biosphere: A plant wax-aerosol proxy approach

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The carbon isotopic ratio (δ^{13}C) of atmospheric CO_2 can help to partition land versus ocean fluxes because terrestrial photosynthesis strongly discriminates against $^{13}CO_2$ whereas ocean uptake does not. This approach requires precise knowledge of large regional scale patterns of carbon isotopic discrimination by terrestrial photosynthesis (Δ) but this has been problematic to quantify given the heterogeneity of terrestrial ecosystems. We are using a proxy approach based upon the δ^{13}C of higher plant-derived leaf wax aerosols in well-mixed continental air masses to scale up terrestrial photosynthetic discrimination (Δ) from the molecular to subcontinental scale. The approach combines the integrating power of atmospheric measurements with the specificity of organic biomarkers to isolate a large-spatially integrated signal of photosynthetic discrimination.

Continuous bulk aerosol measurements (2-week integration period) of wax aerosol molecular and isotopic composition have been made at strategically located sites that receive well-mixed air masses downwind of major ecosystems. High-volume samplers (ocean sectored to prevent local contamination) are used at towers in Bermuda and Barbados. A portable mid-volume unit with an aerosol sampler positioned above the canopy has been used at continental sites (Maine, Alaska, Alberta, Florida, French Guiana). Seasonality in the δ^{13}C of leaf wax aerosols (concentration-weighted C_{24-34} n-alcohols) in the sampled air masses ranges from <2 per mill at Alaska and

Maine sites to approximately 6 per mill at Barbados and Florida. Short-term variability in wax aerosol $\delta^{13}C$ can be attributed to shifting air mass trajectories, for example as observed at the Florida site when north westerlies bringing air masses from mid-latitudes of North America shift to south easterlies bringing subtropical air masses from the Caribbean and North Africa. At Barbados, extremely ^{13}C-enriched wax aerosols are sampled in Oct-Dec during the peak season of C_4 crop harvest/burning in the sub Saharan Africa. The wax aerosol-derived Δ of the air mass "footprints" we have sampled ranges over 10 per mill, from a nearly pure C_4 plant signal of 9 per mill in wintertime air masses arriving at Barbados from North Africa to a nearly pure C_3 plant signal of $>$19 per mill in air masses sampling north temperate ecosystems of North America in late spring. The magnitude and seasonality of the large-scale signal of Δ as estimated by wax aerosol $\delta^{13}C$ compares well with the model estimates of the global distribution of Δ and seasonality in relative contributions of C_3 versus C_4 plant productivity and in the discrimination of C_3 ecosystems.