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Different response of bulk and *n*-alkane δ^{13}C signatures to seasonal shifts in environmental conditions in a temperate coastal ecosystem

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The carbon isotope signal recorded in land plants represents an important reservoir of information for reconstructing climatically driven shifts in plant ecophysiology and biochemistry. Analytical advances have led to widespread usage of compound-specific (CS) carbon isotope analysis of leaf wax biomarkers, such as $n-$alkanes, in addition to traditional bulk isotope methods, to identify shifts in the relative percentage of C_3 and C_4 vegetation contributing to the sedimentary record. Recent studies, however, have extended the application of leaf wax biomarkers, using bulk and $n-$alkane δ^{13}C values interchangeably to derive information about plant-environment relations, both in modern ecosystems and throughout the geological past. Even though previous work on C_3 plants has shown a clear link between climatically influenced plant physiology and bulk δ^{13}C values, further research is needed to establish whether the same link can be seen in leaf wax biomarkers.

To address this question, we collected bulk and $n-$alkane δ^{13}C data from plants growing at Stiffkey marsh on the north Norfolk coast, UK over a period of 15 months. Maximum interspecies variation in weighted average (WA) $n-$alkane δ^{13}C among C_3 species was typically 2-3‰ greater than in bulk. We observed a close correlation in the bulk and WA $n-$alkane δ^{13}C seasonal trends from C_3 grasses and reeds (R^2=0.9, P <0.05). However, for other species (including C_3 and C_4 plants), no statistically significant relationship was observed between their respective bulk and WA $n-$alkane carbon isotope values. This variation in δ^{13}C trends resulted in considerable intra- and inter-species variability (ranging from -4 to -13 per mil) in the offset between bulk and WA $n-$alkane δ^{13}C values. In addition, we identified a positive correlation (R^2=0.7, P <0.05), for all species (with the exception of the evergreen succulent *Suaeda vera*) between the relative abundance of the $n-C_{25}$and $n-C_{27}$ $n-$alkane homologues and $n-C_{29}$ alkane δ^{13}C values.

We explain the discrepancy between bulk and $n-$alkane δ^{13}C signatures by referring to possible interspecies variation in post-photosynthetic carbon isotope fractionation. Our data imply that for some species, seasonal changes in the abundance of $n-$alkane homologues might be an important biochemical process influencing $n-$alkane δ^{13}C signatures. We further theorise that interspecies variation in $n-$alkane δ^{13}C values may arise from biochemical differences in salinity adaptation - in plants adapted to salt stress, production of osmoregulatory solutes (amino acids and/or carbohydrates) may influence the partitioning of pyruvate to fates other than acetyl-CoA, shifting the isotopic composition of lipid biomarkers. Mechanisms controlling metabolic fluxes through these biochemical processes may potentially exert an additional important control over the δ^{13}C signal of lipid biomarkers. We therefore conclude that it may not be valid to use bulk and $n-$alkane δ^{13}C data interchangeably to examine plant-environment interactions. These findings open new avenues for empirical studies to further understand the metabolic processes fractionating carbon during the synthesis of leaf wax lipids, enhancing interpretation of the biomarker signal from the geological record.