Metabolic exchange between pathways for isoprenoid synthesis and implications for biosynthetic hydrogen isotope fractionation

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Hydrogen isotope ratios of plant lipids are used for paleoclimate reconstruction, but are influenced by both source water and biosynthetic processes. Measuring H-2 : H-1 ratios of multiple compounds produced by different pathways could allow these effects to be separated, but hydrogen isotope fractionations during isoprenoid biosynthesis remain poorly constrained. To investigate how hydrogen isotope fractionation during isoprenoid biosynthesis is influenced by molecular exchange between the cytosolic and plastidial production pathways, we paired position-specific C-13-pyruvate labeling with hydrogen isotope measurements of lipids in Pachira aquatica saplings. We find that acetogenic compounds primarily incorporated carbon from (13)C2-pyruvate, whereas isoprenoids incorporated (13)C1- and (13)C2-pyruvate equally. This indicates that cytosolic pyruvate is primarily introduced into plastidial isoprenoids via glyceraldehyde 3-phosphate and that plastidial isoprenoid intermediates are incorporated into cytosolic isoprenoids. Probably as a result of the large differences in hydrogen isotope fractionation between plastidial and cytosolic isoprenoid pathways, sterols from P. aquatica are at least 50 parts per thousand less H-2-enriched relative to phytol than sterols in other plants. These results provide the first experimental evidence that incorporation of plastidial intermediates reduces H-2/H-1 ratios of sterols. This suggests that relative offsets between the H-2 : H-1 ratios of sterols and phytol can trace exchange between the two isoprenoid synthesis pathways.
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