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## The dominant environmental driver of leaf water stable isotope enrichment differs for H-2 compared to O-18

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Several important isotopic biomarkers derive at least part of their signal from the stable isotope composition of leaf water (e.g., leaf wax  $\delta^2\text{H}$ , cellulose  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$ , lignin  $\delta^{18}\text{O}$ ). In order to interpret these isotopic proxies, it is therefore helpful to know which environmental variable most strongly controls a given leaf water stable isotope signal. We collated observations of the stable isotope compositions of leaf water, xylem water, and atmospheric vapour, along with air temperature and relative humidity, to test whether the dominant driver of leaf water  $^2\text{H}$  concentration could differ from that of  $^{18}\text{O}$  concentration. Our dataset comprises 690 observations from 35 sites with broad geographical coverage. We limited our analysis to daytime observations, when the photosynthetic processes that incorporate the leaf water isotopic signal primarily take place. The Craig-Gordon equation was generally a good predictor for daytime bulk leaf water stable isotope composition for both  $\delta^2\text{H}$  ( $R^2=0.86$ ,  $p<0.001$ ) and  $\delta^{18}\text{O}$  ( $R^2=0.63$ ,  $p<0.001$ ). It showed about 10% admixture of source water was caused by unenriched water pools such as leaf veins or the Péclet effect. Solving the Craig-Gordon equation requires knowledge of relative humidity, air temperature, and the stable isotope compositions of source water and atmospheric vapour. However, it is not possible to invert the Craig-Gordon equation to solve for one of these parameters unless the others are known. Here we show that the two isotopic signals of  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  are predominantly driven by different environmental variables: leaf water  $\delta^2\text{H}$  correlated most strongly with the  $\delta^2\text{H}$  of source water ( $R^2=0.68$ ,  $p<0.001$ ) and atmospheric vapour ( $R^2=0.63$ ,  $p<0.001$ ), whereas leaf water  $\delta^{18}\text{O}$  correlated most strongly with air relative humidity ( $R^2=0.46$ ,  $p<0.001$ ). We conclude that these two isotopic signals of leaf water are not simply mirror images of the same environmental information, but carry distinct signals of different climate factors, with crucial implications for the interpretation of downstream isotopic biomarkers.

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