

The supporting information demonstrates the derivation of $(\Delta S/S_0)^{lab}$ for the HFP2-¹⁴AAG and HFP4-¹⁴A samples. The $(\Delta S/S_0)^{lab}$ refers to the signals expected for the labeled ¹³COs and is calculated by removing natural abundance (*na*) ¹³CO contributions from $(\Delta S/S_0)^{exp}$:

$$\left(\frac{\Delta S}{S_0}\right)^{exp} = \frac{S_0^{lab} + S_0^{na} - S_1^{lab} - S_1^{na}}{S_0^{lab} + S_0^{na}} = 1 - \frac{S_1^{lab}}{S_0^{lab} + S_0^{na}} - \frac{S_1^{na}}{S_0^{lab} + S_0^{na}} \quad (1)$$

Multiplication of the left-most and right-most sides of Eq. 1 by $(S_0^{lab} + S_0^{na})/S_0^{lab}$ is followed by algebraic manipulation:

$$\left[\left(1 + \frac{S_0^{na}}{S_0^{lab}}\right) \times \left(\frac{\Delta S}{S_0}\right)^{exp}\right] = \left(\frac{\Delta S}{S_0}\right)^{lab} + \frac{(\Delta S^{na})}{S_0^{lab}} \quad (2)$$

Multiplication of the right-most term by (S_0^{na}/S_0^{na}) is followed by algebraic manipulation to yield Eq. 2 from the main text:

$$\left(\frac{\Delta S}{S_0}\right)^{lab} = \left[\left(1 + \frac{S_0^{na}}{S_0^{lab}}\right) \times \left(\frac{\Delta S}{S_0}\right)^{exp}\right] - \left[\left(\frac{S_0^{na}}{S_0^{lab}}\right) \times \left(\frac{\Delta S}{S_0}\right)^{na}\right] \quad (3)$$

with (S_0^{na}/S_0^{lab}) calculated from the numbers of natural abundance and labeled ¹³COs in the sample and $(\Delta S/S_0)^{na}$ calculated as the average of $(\Delta S/S_0)^{exp}$ for the HFP2-⁵GAL, HFP3-⁸FLG, and HFP2-¹¹FLG samples. Although the latter calculation is an approximation, uncertainties in $(\Delta S/S_0)^{na}$ have a relatively small impact on the uncertainty of $(\Delta S/S_0)^{lab}$. For example, consider the spectra for the HFP2-¹⁴AAG/PC:PG sample at $\tau = 24$ ms. The values of (S_0^{na}/S_0^{lab}) , $(\Delta S/S_0)^{exp}$, and $(\Delta S/S_0)^{na}$ are 0.084, 0.419 ± 0.014 , and 0.134, respectively, and result in $(\Delta S/S_0)^{lab} = 0.443 \pm 0.015$. If $(\Delta S/S_0)^{na}$ were 0.0 or 0.25, $(\Delta S/S_0)^{lab}$ would be 0.454 or 0.433, and are within the experimental uncertainty of the reported $(\Delta S/S_0)^{lab}$.