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A comprehensive model for COS isotope discrimination during leaf COS uptake

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Anthropogenically emitted CO_2 is warming the earth's climate to temperatures that already exceed pre-industrial levels by more than 1.2 ^{o}C. Terrestrial vegetation has slowed the rate of climate change by removing part of this anthropogenic emission. Accurate estimations of the present and future terrestrial carbon sink are still needed for forecasting climate and for informing policies for climate stabilization. This requires precise knowledge of the photosynthetic C uptake over land (gross primary production, GPP), independently of the C released through plant and soil respiration. The gas carbonyl sulfide (COS) has emerged as a promising tracer for GPP. This is because both CO_2 and COS are a substrate for carbonic anhydrase (CA), the first enzyme involved in photosynthesis, so that the uptake by foliage of COS and CO_2 often covaries. Estimating GPP from COS measurements and atmospheric budgets also requires quantifying ocean and industrial COS sources, which is challenging. Isotopic constrained COS tropospheric mass balances can help quantify the relative contribution of these sources if the isotope discrimination during COS uptake by terrestrial vegetation (the main COS sink) is known. However, little is known about plant-atmosphere COS isotope exchange; measurements are challenging and theory to interpret these measurements is limited. Herein, we present a new comprehensive model for discrimination during COS uptake by plants ($\Delta^{34}S$) and use it to revisit existing COS isotope datasets and atmospheric budgets. Our $\Delta^{34}S$ model expands Davidson *et al*. (2022) pioneer framework by accounting for leaf COS production. By analogy with the well-established model for photosynthetic discrimination against $^{13}CO_2$, Davidson *et al*. $\Delta^{34}S$ model stated that COS discrimination occurs as COS diffuses into the leaf and binds to CA. Leaf COS emission was not considered, although it has been reported in species ranging from bryophytes to wheat and trees. Because it is uncertain *where* these emissions occur, we tested different leaf-level COS emission scenarios - including zero emissions - in various leaf compartments (cuticle, intercellular space, cytosol), alone or in combination. We used this comprehensive model to generate predictions for $\Delta^{34}S$ in C_3 and C_4 species and discussed implications for determining a global plant uptake fractionation factor. Our mechanistic model provides a framework to interpret vegetation-atmosphere COS isotope exchange that can prove useful to improve COS uptake-based GPP estimates and our understanding of plant function, especially when combined with other isotopes (C, O, H).