

Reactive-transport models and isotopic tracers reveal the spatial pattern of riverine CO₂ emissions and suggest cooperative inorganic-organic carbon cycling

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Some fraction of the dissolved inorganic carbon (DIC) transported by rivers is released into the atmosphere as carbon dioxide during transit from source to sink. Besides being a substantial flux of carbon, gas evasion can alter the isotopic composition of DIC and complicate quantifications of chemical weathering. Calculating the evaded portion of DIC is complicated by the need to constrain chemical and physical parameters along an entire stream network using a limited number of point measurements. Reactive transport models are a promising tool to interpolate between point measurements, but it remains unclear whether riverine observations alone can accurately inform models or if additional measurements of catchment carbon sources are required to estimate gas evasion.

We found that spatially distributed observations of riverine carbonate chemistry, $\delta^{13}\text{C}$ -DIC, and ^{222}Rn were sufficient to constrain a reactive transport model of gas evasion in a silicate-dominated catchment in Oregon (Little Deschutes River). This was demonstrated by comparing the model results to independent measurements of soil gas and groundwater chemistry. Instead of relying on global parameterizations for the dependence of the gas exchange coefficient on channel hydraulics, our approach enabled us to develop a site-specific scaling relationship. Using complementary measurements of solute concentrations, we determined that nearly all the DIC at our study site was sourced from organic matter respiration with little to no contribution from carbonate weathering. This allowed us to use our model and additional radiocarbon data to constrain the relationship between silicate weathering, soil respiration, floodplain carbon storage, and CO₂ evasion. Specifically, we found that the respiration of young organic matter efficiently generates alkalinity via silicate weathering such that only 20% of the total DIC flux is lost via gas evasion. The products of this rapid silicate weathering, secondary clay minerals, in turn allow for the storage of organic matter in floodplain soils for millennia, highlighting a