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Developing Sulfur Radio- and Stable Isotope Approaches to Determine Changes to the Terrestrial Sulfur Cycle

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Sulfur (S) is a critical element for plant growth. As atmospheric S deposition has declined in response to air quality regulation in the United States (U.S.) and Europe, there has been an increase in S fertilizer applications reported in many crop systems. This increase in S use over large agricultural regions represents humans' new manipulation of the global S cycle. Given that excess S can cause soil acidification and mobilization of heavy metals in ecosystems, it is critical to develop methods to trace the "fingerprint" of agricultural S through watersheds, quantify S forms and transformations in soils and surface waters, and determine the consequences of its use. We present a suite of studies that have adapted radio- and stable isotope methods from the marine literature for studying changes to the terrestrial S cycle. We focus our research in the Napa Valley, California (U.S.) winegrowing region, where winegrowers apply elemental S to control powdery mildew. We used a combination of Fourier transform ion cyclotron resonance mass spectrometry, $\delta^{34}S$-dissolved organic S (DOS), and S X-ray absorption spectroscopy to characterize DOS in soil porewater and surface water samples from vineyards (plus S) and forest/grassland areas (no S). Vineyard soil porewater dissolved organic matter samples had two-fold higher S-content compared to forest/grasslands and had unique $CHOS_2$ chemical formulas—the latter were also found in vineyard tributaries and the Napa River. We also used the radioisotope, $^{35}S\text{-}SO_4^{2-}$, to quantify sulfate reduction rates (SRR) within vineyard and forest/grassland soils paired with silver films buried to identify locations of sulfate reduction in soil profiles. These data demonstrate that sulfate reduction occurs in vineyards during saturated periods in the wet (dormant) season; within soils, it occurs in small aggregates, and, thus, SRR are low. In vineyard drainages, SRR are 6 orders of magnitude higher than in vineyard soils and occur in distinct subsurface zones. These data indicate the utility of S isotopes for tracing new patterns and transformation pathways associated with the human manipulated S cycle.