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Sulfur speciation during low- to moderate-intensity bushfires: a sulfur K-edge XANES investigation

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Climate change is predicted to increase the frequency, severity, and spatial extent of bushfires. Fire is now recognised as a potent driver of nutrient and contaminant redistribution at local to global scales, yet its influence on arsenic (As), iron (Fe), and sulfur (S) geochemistry remains poorly resolved. Sulfur is highly redox-sensitive, and bushfire combustion processes can induce rapid oxidation, reduction, and phase transformation of Fe–As minerals, thereby influencing S–As–Fe environmental mobility. Sulfur is also a central component of acid sulfate systems, including acid mine drainage (AMD) and acid sulfate soils (ASS), where it plays a critical role in acid generation and Fe and As speciation. Despite this, sulfur redox transformations during heating events remain largely unexplored. This study investigates sulfur transformations during thermal heating of soils containing Fe–As–S phases under bushfire-relevant conditions, and evaluates how organic matter composition influences sulfur redox pathways and S–Fe coordination during moderate-intensity fires. Twenty-four soils from seven sites affected by AMD and ASS were heated at 0, 175, 200, 250, 300, 350, and 400°C. Selected soils were also examined for time-dependent changes. Sulfur K-edge XANES was collected in all treatment to resolve sulfur speciation. Iron and As K-edge X-ray absorption spectroscopy were also collected to selected samples. Sulfur speciation varied substantially between soils. Samples containing minimal organic sulfur were designated as “mineral” soils. In these soils, speciation changes were limited until 400°C, where structural sulfate (e.g., gypsum or jarosite) lost crystallinity and shifted toward less-ordered or adsorbed sulfate forms. In contrast, samples rich in poorly crystalline phases, including schwertmannite and sulfate-substituted amorphous ferric arsenate, showed little change in sulfate coordination across the heating range. Organic sulfur species in the remaining soils were dominated by cystine, cysteine, and sulfonate groups. Cystine and sulfonate species were particularly temperature-sensitive, progressively oxidising to sulfate from 175°C, with near-complete conversion at 400°C. However, oxidation rates were soil-dependent. This work provides the first known systematic spectroscopic evaluation of sulfur redox transformations in AMD- and ASS-affected soils under bushfire-relevant heating regimes.