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Sulfur Redox Status in Peatland Soil and Outflow Waters Diverges with Climate Warming

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Boreal peatlands are important reservoirs of carbon and other elements. Increasing temperatures and more variable precipitation patterns alter oxidation-reduction (redox) conditions and fluxes of atmospheric and aquatic pollutants from peatlands. Here, we measure the effect of warming and elevated carbon dioxide on the speciation of sulfur in boreal peatland soil and outflow water over three years. In a whole-ecosystem warming experiment, with temperature levels of +0 °C (control), +2.25 °C, +4.5 °C, +6.75 °C, and +9 °C above ambient, water table height was negatively correlated with warming. Warming was correlated with changes in the size of sulfur pools: sulfur content (weight%) decreased in soils and sulfate concentrations increased in outflow. Reflecting the warmer and drier conditions, the percentage of oxidized sulfur in soil, as measured by X-ray absorption near edge structure (XANES) spectroscopy, increased with warming. Sulfur speciation in soil showed increases in ester-sulfate ($R\text{-}O\text{-}SO_3^-$) content at the expense of organic disulfide (R-S-S-R') content. In contrast to the soil, the percentage of oxidized sulfur decreased in outflow with warming. The changes in sulfur speciation in outflow were characterized by increased organic monosulfide (R-S-R', R-S-H) content at the expense of ester-sulfate. Overall, the peatland sulfur pools became more oxidized in the soil and more chemically reduced in the outflow water in response to soil and air warming. The connection between these opposite redox trends is likely enhanced microbial activity in porewaters and outflow with warming. We observed that ester-sulfate partitioned from soil to outflow waters during heavy rainfall periods, making it more available for microbial sulfur-reduction. Notably, no significant effect of elevated carbon dioxide on sulfur pools was detected. Our findings are consistent with a net export of organic sulfur from the peatland to receiving surface waters. The overall loss of sulfur from this peatland is consistent with enhanced decomposition and increased plant available nutrients reported previously for this whole-ecosystem warming experiment. Warming-induced changes to sulfur pools in peatlands affect the fluxes of other constituents, such as the pollutant methylmercury, with downstream consequences on water quality.