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Accelerating sulfide oxidation in the Mackenzie River catchment, Canada

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Rivers integrate signals of physical and chemical processes occurring across catchment landscapes. Under a warming climate in the Arctic, river geochemistry changes can be indicative of changing rock weathering rates, hydrological pathways and the multiple consequences of permafrost thaw. In the Mackenzie River, northwestern Canada, sulfuric acid-driven mineral weathering has been identified as an important but understudied component of the regional carbon cycle. Shale lithologies across the catchment contain sulfides that can be oxidised to produce dissolved sulfate (SO_4^{2-}). Sulfuric acid-driven dissolution of carbonate minerals releases CO_2 to the atmosphere and a growing body of work suggests that it has the potential to have a positive feedback on climate warming. Trends of increasing in sulfate export have been identified for major tributaries of the Mackenzie and have been linked to accelerating thermokarst processes [1,2]. In alpine catchments, sulfate export has been associated with warming across multiple sites [3] and CO_2 export from oxidative weathering has been linked directly to temperature at the site scale [4]. The response of sulfide oxidation in the Mackenzie to temperature, however, has not yet been quantified.

We analyse a multi-decadal dataset (1970 – 2020) of river geochemistry and discharge from a suite of nested sub-catchments across the Mackenzie watershed. Studied sub-catchments range in size and elevation, drain regionally representative lithologies (shale, carbonates, shield) and span continuous to sporadic permafrost zones. Using a gridded climate reanalysis dataset for the corresponding time period, we establish the relationship between catchment-scale temperature change and variability in sulfate flux. We then use geospatial analyses to link regional differences in the direction and magnitude of these changes to catchment characteristics. This dataset allows for both a spatial and temporal assessment of the controls on sulfide oxidation in the Mackenzie on seasonal to decadal timescales and enables the associated carbon budget to be estimated.

[1] Tank et al. (2016) *ERL* 11, 054015
[2] Zolkos et al. (2018) *GRL* 45, 9623-9632
[3] Crawford et al. (2019) *ERL* 14, 124092
[4] Soulet et al. (2021) *Nature geoscience* 14, 665-671