

## **Erosion and weathering drive net CO<sub>2</sub> drawdown in the Mackenzie River Basin over geological time**

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Erosion and weathering transfer carbon between the atmosphere and the lithosphere, thereby acting to modify Earth's long-term climate. Over millions of years, atmospheric carbon dioxide (CO<sub>2</sub>) is sequestered during the weathering of silicate minerals by carbonic acid, coupled to carbonate formation, along with the erosion of biospheric organic carbon and its burial in sediments. However, erosion and weathering also act together to release CO<sub>2</sub> from the lithosphere. Erosion enhances the rate of oxidative weathering of organic carbon in rocks (petrogenic OC, OC<sub>petro</sub>), which is a major CO<sub>2</sub> source over geological time. In addition, the oxidation of sulfide minerals can produce sulfuric acid that weathers carbonate minerals and results in transient CO<sub>2</sub> release. Although these sources and sinks of CO<sub>2</sub> are well recognised, limited case studies exist where they have been measured alongside each other.

Here, we calculate the geological carbon budget accompanying weathering and erosion in the Mackenzie River Basin, Canada. The silicate weathering rate, carbonate weathering rate by sulfuric acid and the sedimentary burial of biospheric organic carbon have been constrained by prior work to be 0.4 tC km<sup>-2</sup> yr<sup>-1</sup>, 0.7 tC km<sup>-2</sup> yr<sup>-1</sup> and 2 tC km<sup>-2</sup> yr<sup>-1</sup>, respectively. Closing the long-term CO<sub>2</sub> budget therefore requires quantification of the OC<sub>petro</sub> oxidation rate. To do this, we use dissolved rhenium (Re) flux as a proxy for OC<sub>petro</sub> weathering with samples collected from 2009 to 2013.

In the Mackenzie Basin, over 90% of dissolved Re appears to be derived from OC<sub>petro</sub>; this is according to [Re]/[Na] and [Re]/[SO<sub>4</sub><sup>2-</sup>] ratios in OC<sub>petro</sub>, sulphide and silicate mineral endmembers. Using estimates of the coupled mobility of Re and CO<sub>2</sub> during OC<sub>petro</sub> weathering, we constrain the CO<sub>2</sub> flux from OC<sub>petro</sub> weathering to be ~0.3 tC km<sup>-2</sup> yr<sup>-1</sup>. Consequently, OC<sub>petro</sub> oxidation cannot negate the large CO<sub>2</sub> sink driven by biospheric organic carbon erosion, and under the present climate state the Mackenzie Basin is a net CO<sub>2</sub> sink.