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Release of aged CH_4 from Arctic Rivers: Isotopic insight on sources and formation pathways

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Rivers are now recognised as a major pathway of methane (CH_4) from terrestrial organic carbon stores to the atmosphere, releasing a combined 27.9 $TgCH_4$ yr^{-1}, which is similar to other freshwater systems. Despite this recognition, we lack observations of river CH_4 concentrations and fluxes in many regions. There are even fewer measurements of river CH_4 isotopic composition ($\delta^{13}C$, $\delta^{2}D$ and $F^{14}C$) which allow us to constrain its source, biogeochemical pathways and the consequences of its emission. The major Arctic Rivers and their deltas are of particular relevance, where ongoing rapid warming is leading to gradual and abrupt thaw of permafrost, which collectively stores 1035±150 peta-grams of carbon (Pg C) in the upper 3 meters. With limited sampling of river CH_4 in Arctic rivers we lack a clear understanding whether river CH_4 emissions could contribute to the permafrost carbon climate feedback.

Here, we address this research gap in the Mackenzie River, its major Arctic tributaries, and delta region. At these sites, we measure dissolved CH_4 and carbon dioxide (CO_2) concentrations and river CH_4 and CO_2 emissions during the fall shoulder season and subsequent ice break up and peak flow. We collect samples for radiocarbon (^{14}C) analyses and stable isotopes using modified novel sampling techniques. We find the age of riverine dissolved CH_4 ranged from 'modern' to 2012±77 ^{14}C years BP (Fraction Modern ($F^{14}C_{CH4}$) of 1.02 to 0.77). These ages suggest a dominance of biogenic CH_4, rather than thermogenic CH_4 ($F^{14}C$ = 0). A distinct pattern in the evolution of ages, stable isotope signatures provide evidence for seasonal shifts in the dominance of methane formation pathways. We find elevated CH_4 fluxes in regions of sediment accumulation in tributaries, and find CH_4 fluxes increase markedly as the river system enters the delta. We estimate a diffusive CH_4 flux from river channels in the delta which is comparable to the better studied flux of CH_4 from lakes, but note that the riverine CH_4 is from older C stores. The CH_4 release from sediment-rich hotspots of Arctic Rivers could be a significant source of aged C flux which exacerbates the permafrost carbon cycle feedback.