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A radiocarbon-based inventory of methane and inorganic carbon dissolved in surface lake waters in arctic Alaska, USA

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Major uncertainties in land-atmosphere carbon (C) exchange in the rapidly warming and wetting Arctic are 1) the magnitude and timing of net losses of ancient permafrost C to the atmosphere and 2) the relative changes of C exchange as carbon dioxide (CO_2) or the more powerful greenhouse gas methane (CH_4). For CH_4, the role of diffusive fluxes versus plant-mediated and ebullition fluxes is poorly constrained. Radiocarbon (^{14}C) is a unique tracer for distinguishing ancient permafrost C from C rapidly cycling between the land and atmosphere. In addition, stable isotope ratios ($^{13}C/^{12}C$ and D/H) provide insight to trace gas production and consumption pathways. Previous measurements, however, have focused on CH4 from ebullition fluxes due to technical and logistical challenges in ^{14}C-CH_4 analysis.
We quantified the ^{14}C content and δ13C signatures of dissolved CH_4 and DIC in lake surface waters along a north-south transect on the North Slope of Alaska, USA (69.9°N to 71.28°N, -156.12°W to -156.32°W). Samples were collected at the end of winter before ice break-up (April 2013) and during summer (August 2012 & 2013) in 1 L bottles. A subset of samples was also analyzed for CH_4 and CO_2 concentrations and stable isotope ratios by the Circumarctic Lakes Observation Network (CALON). In addition, in August 2013, we measured the ^{14}C content and $\delta^{13}C$ ratios of lake-atmosphere CH_4 and CO_2 exchange near Barrow, AK, USA (71°N, -156°W). We obtained dissolved CH_4 and CO_2 sufficient for ^{14}C analysis from lakes with concentrations as low as 0.01 mg C /L) using a novel, in situ preconcentration method (liqui-cel, Membrana). And, we measured and collected isoflux samples of simulated, near-shore ebulltion-derived CH_4 and CO_2 using floating headspace chambers.
Isotope samples were processed using a novel, flow-through vacuum line and analyzed at the KCCAMS facility at the University of California, Irvine, USA with accelerator (0.5MV 1.5SDH-2, National Electrostatics Corporation) and isotope-ratio mass spectrometry (Gas Bench coupled with Finnigan DeltaPlus, Thermo).

Preliminary data indicate that along the transect summertime DIC concentrations range from 112.4 μM to 1619.8 μM, with a ^{14}C content of 0.87 FM to modern and $\delta^{13}C$ ratios of -23.5 to +3.0 ‰. We found no relationship between ^{14}C content and latitude, however regional clusters of lakes had similar isotope signatures.
Summertime concentrations of dissolved CH_4 were mostly below the analytical limit for ^{14}C analysis, except for one modern sample.

Within the Barrow region, summertime dissolved CH_4 was depleted in ^{14}C in lakes with surface areas $\geq$8 ha, but modern in ponds <8 ha (FM 0.92±0.09 vs. 1.02±0.02). In lakes, ebullition-derived ^{14}C-CH_4 fluxes had similar ^{14}C contents as dissolved CH_4 (FM 0.97±0.05 vs. 0.92±0.09). However, in ponds, ebullition-derived CH_4 was ^{14}C-depleted relative to dissolved CH_4 (FM 0.98±0.05 vs. 1.02±0.02). Dissolved CH_4 originated from older sources than dissolved CO_2, especially in lakes.