Data was collected at Brownie Lake in Minneapolis, MN, USA and at Canyon Lake in the Upper Peninsula of MI, USA. The methane flux, or the amount of methane entering the atmosphere over a given area per time, was assessed at Brownie Lake and Canyon Lake. Sampling of Brownie Lake during geochemical characterization revealed that the methane flux out of the lake was much higher than Canyon Lake, as well as other ferruginous meromictic lakes used as geochemical analogs for early Earth’s ferruginous oceans. The dataset here was used to discern why the methane flux out of Brownie Lake was high. Brownie Lake was sampled in May, July and September of 2017 and June 2018. Canyon Lake was sampled in June and September 2017 and May 2018. We used various limnological probes and sensors (LDO sensor, Hydrolab multiprobe) to collect water column profiles (temperature, dissolved oxygen, specific conductance, chlorophyll a, pH). We also analyzed water samples (cations, anions, CH4, DIC) using ion chromatography and ICP-MS and measured isotopes (CH4, DIC) utilizing isotope ratio mass spectrometry. The methane flux was constrained in two ways: taking direct samples from the surface of each lake using floating static flux chambers, which were measured by gas chromatography, and estimated from geochemical reaction-transport modeling based on the diffusional profiles of methane and other dissolved species throughout the water column.