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The behavior of methane clumped isotopologues in a wetland-landfill-air mixing system

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Ever since first methane clumped isotopologue measurements of air thought possible (Haghnegahdar et al., 2021), they were expected to provide unique constraints on the mixing of methane sources with air. A few studies (Sun et al., 2022; Fernandez et al., 2022; Haghnegahdar et al., 2022) shown that clumped isotopologues can provide information about mixing similar to that provided by carbon and hydrogen isotopes.

We monitored methane concentrations, carbon and hydrogen isotopes, and clumped isotopologues of morning air on a monthly schedule at Patuxent river wetlands park. This location is associated with the scuttling of Chesapeake Flotilla in 1814. Additional samples were collected upstream and downstream along Patuxent River. Samples were collected before sunrise to capture methane accumulated overnight before atmosphere temperature inversion was dissipated by ground heating up.

The most significant sources at the site are local wetlands, Washington DC methane dome and, when winds are from northeast, Brown Station landfill. Brown Station generates high methane concentration plumes that can be detected for miles. A smaller landfill exists about 2 miles north and a wastewater treatment plant with an associated composting facility is located about 2 miles southeast. It is not yet clear if these other sites are significant contributors of methane to the site.

Measurement results and mixing models show that for a specific mixing system involving three (or mainly three) endmembers, clumped isotopologues present a narrow and twisted knot-shaped mixing grid with highly dense grids at both ends (Fig 1). Clumped isotopologue signals of air will not be largely altered by mixing with up to 30% mixing ratio of sources and mean that considering only clumped isotopologues will not be suitable for distinguishing mixing relationships. However, this behavior of clumped isotopologues provides other information. We can use methane concentration, carbon and hydrogen isotopes to get the endmember compositions (Fig 2), and use these constraints to derive the clumped isotopologue mixing grids (Fig 1). Any deviations from the mixing grids may show the source variability itself (especially air). Just like what happens during a total solar eclipse, the corona that was originally overwhelmed by photosphere is finally revealed.

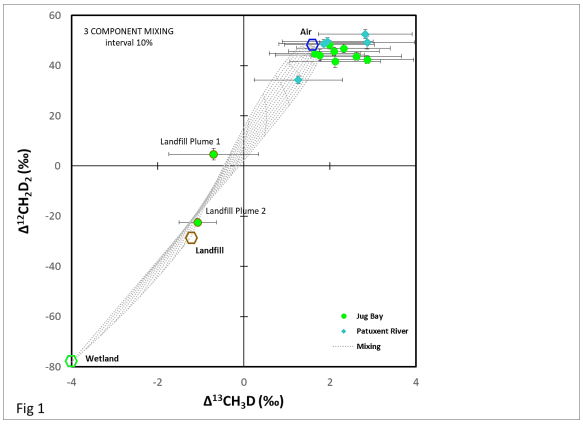

Fig 1

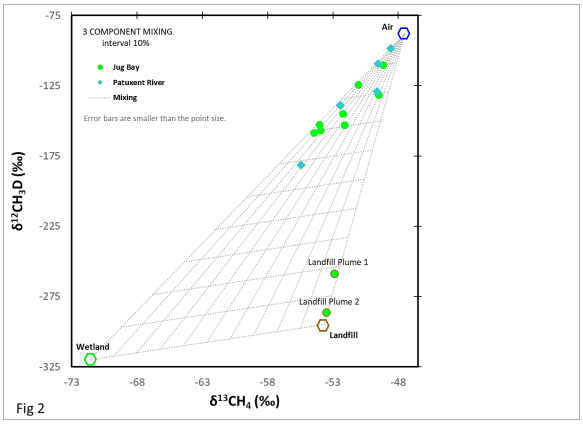

Fig 2