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Origins of methane within diverse geothermal systems

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Methane is the second most important anthropogenic greenhouse gas in Earth's atmosphere, yet there remains great uncertainty in its global budget, in particular with regards to the potential hidden source(s) of methane [1]. It is therefore important to determine atmospheric methane fluxes for understanding past and future climate, and how gas releases may change with changes to Earth's climate. Natural methane emissions include those from volcanism, hydrothermal releases, and forest fires. Within volcanic and geothermal systems there can be multiple sources of methane, including biotic, abiotic and thermogenic [2] and it is important to understand the role these different processes play in controlling the flux of methane to the atmosphere to better constrain the global methane budget.

The sources of methane are typically determined using 'bulk' isotopic measurements of methane $\delta^{13}C$ and δD [3,4], yet there is significant overlap in the composition of the different sources making it difficult to determine the source. However, as methane forms there is strong isotopic discrimination between doubly substituted "clumped" isotopologues ($\Delta^{13}CH_3D$ and $\Delta^{12}CH_2D_2$) allowing for the robust discrimination between abiotic, biotic and thermogenic methane. This makes the doubly substituted isotopologues of methane an important tool for characterising the sources and sinks of methane.

The diverse nature of geothermal systems in Yellowstone National Park and across Italian Peninsula Volcanic Province make them ideal locations to understand methane sources within geothermal areas. Here, we compare methane isotopologues and noble gas isotopes in three samples from Yellowstone National Park, one sample from the Yellowstone hotspot track and four samples from across Italy, to investigate the different sources of methane across a geothermal region. We find a wide range in helium isotopes (from radiogenic to plume-like) and clumped methane isotopes. We find a correlation between clumped methane isotopes and helium isotopes, suggesting there is a relationship between the amount of mantle derived volatiles and methane within the system and use these data to help identify the processes controlling methane generation and migration within hydrothermal systems.

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