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Hydrocarbon cycling in Tokamachi mud volcano, Niigata, Japan

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Mud volcanoes and gas seepages represent a significant natural source of hydrocarbons to the atmosphere [1], a window to subsurface life, as well as potential analogs for Mars [2]. Although previous studies have been conducted to quantify hydrocarbon fluxes in geologic sources, the different pathways involved in the production and recycling of hydrocarbons in the subsurface remains poorly constrained. This mostly stems from the difficulty in detecting and differentiating processes using conventional isotopic methods. Here we study the Tokamachi mud volcano as a model geological setting for hydrocarbon-based subsurface systems through the use of incubation techniques and recent advances in stable isotope analysis. We take advantage of the several seepages in the area showing different fluid compositions, which gives a good opportunity to study fluids with potentially different histories. The natural gases comprise mainly of methane (85.7-97.3%) and CO_2(7.3-21.7%) followed by minor percentages of C_{2+} hydrocarbons. Bulk isotope analysis of hydrocarbons agrees with previously established thermogenic origin of gases ($\delta13C_{CH4}$ -40.1 to -30.3 ‰), as well as the presence of biodegradation based on enriched 13C of propane and CO_2 [3]. However, this biodegradation and its extent has yet to be confirmed. Here, we use position-specific isotope analyses (PSIA) of propane and butane which have recently been shown to provide a quantitative estimation of the extent of hydrocarbon biodegradation in natural gas basins [4]. The analyses of gases from multiple seeps show increasing $\delta13C_{CH4}$ with increasing degrees of biodegradation, which could be explained by secondary methanogenesis using products of hydrocarbon degradation. This link between degradation and methanogenesis is further supported by CH_4 data from cultivation experiments using indigenous mud and different substrates. Moreover, preliminary results from 16S rRNA amplicon sequencing and carbon isotope analysis of our methanogenic enrichment cultures show that the likely pathways of microbial CH_4 formation in Tokamachi are hydrogenotrophic and acetoclastic methanogenesis. Combining data from enrichment cultures, PSIA, 16S rRNA amplicon sequencing, and thermodynamic calculations, we present a model constraining the sources and fates of hydrocarbons in the Tokamachi mud volcano.

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