

Origin of ultramafic CH₄: the CH₄-H₂-H₂O hydrogen isotope systematics of the Hakuba Happo hot spring

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Ultramafic-hosted hydrothermal fluids are characterized by high concentration of H₂ derived from serpentinization. Ultramafic rock is rare in the surface of the Earth today, but is likely to have been an abundant component of the early crust due to higher potential mantle temperature. Ultramafic-hosted hydrothermal ecosystem has attracted attention as a potential habitat of the Earth's earliest microbial community because the earliest chemolithoautotrophs may have utilized molecular H₂ as an electron donor. Along with hydrogen, high concentrations of methane and hydrocarbons have been reported in ultramafic hydrothermal fluid. They are usually ¹³C-rich compare to microbially-produced methane, and could have been synthesized by abiotic reactions. However, origin of the methane in the ultramafic hydrothermal system is not clearly understood yet. In this study, we collected fluid samples from ultramafic rocks in Hakuba Happo hot spring, and conducted chemical and C-H isotopic analyses of the CH₄ as well as compounds possibly involved in the methane formation reaction in fluids. Samples from Happo hot spring located on the serpentinite body were directly collected from two drilling wells (Happo #1 and Happo #3). The hot spring water is strongly alkaline (pH>10) and rich in H₂ (201~664 umol/L) and CH₄ (124~201 umol/L). These chemistries were typical of fluids associated with ultramafic rock. Even lower temperature regime, H₂ in Hakuba Happo is derived from serpentinization. We measured the concentrations of dissolved gas and the hydrogen and carbon isotope compositions of H₂, CH₄, CO₂ and H₂O. Hydrogen isotope compositions for Happo #1 were dD-H₂= -700 permil, dD-CH₄= -210 permil, dD-H₂O= -84.5 permil, and those for Happo #3 were dD-H₂= -710 permil, dD-CH₄= -300 permil, dD-H₂O= -84.2 permil. Hydrogen and water at Happo#1 had similar dD values to those at Happo#3, but methane from Happo#1 was approximately 80 permil enriched in deuterium relative to Happo #3. On the other hand, carbon isotope compositions of methane from Happo#1 and Happo#3 were d¹³C= -34.5 permil and -33.9 permil, respectively, and there was almost no difference. The CH₄-H₂-H₂O hydrogen isotope systematics suggests that the most likely production process of Happo #1 methane is the olivine hydration with carbon source, and that biological methane contributes to Happo #3 methane.

Keywords: ultramafic-hosted hydrothermal system, origin of CH₄, hyperalkaline, hydrogen isotope composition