Origin of methane in Hakuba Happo serpentinite-hosted hot spring: ^{14}C and noble gas study

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High concentration of methane (CH_4) and higher molecular weight hydrocarbons (C_{2+}) are commonly observed in water-rock interaction systems associated with serpentinization (e.g., Schrenk et al., 2013; Etiope and Sherwood-Lollar, 2013). Previous studies have suggested that stable hydrogen and carbon isotopic compositions of hydrocarbons observed at some serpentinization sites are compatible with abiotic formation via polymerization process (Proskurowski et al., 2008; Suda et al., 2017). However, time and space for the formation of hydrocarbons are still poorly constrained; when and where hydrocarbons are produced? In this study, we report the results of radiocarbon (^{14}C) and noble gas analyses for serpentinized-hosted hot spring in Hakuba Happo, Japan.

The hyperalkaline water (pH>10) with moderate temperature (ca. 50°C) is pumped from two borehole wells. Water chemistry and volatile component are controlled primarily by serpentinization reaction (Homma and Tsukahara, 2008; Suda et al., 2014). Samples were collected in 2015-2018. The ^{14}C measurement reveals that CH_4 at Hakuba Happo is ^{14}C dead, i.e., the ^{14}C ages of CH_4 is at least approximately 50,000 years before present. On the other hand, calcium carbonate scale precipitating on steel piping contains modern carbon. Radiocarbon evidence rules out a groundwater bicarbonate as the carbon source of CH_4 at Hakuba Happo hot spring system. The high helium isotope ratio ($^3He/^4He$) was observed in both well sites. This indicates that the mantle-derived helium component is carried to the Hakuba Happo hot spring system. For one site, relative contribution of air, mantle and crustal components are estimated to be 47%, 31% and 22%, respectively. For the other site, 69%, 24% and 8%, respectively. The radiocarbon and helium isotope data suggest that the requisite carbon source for the Hakuba Happo CH_4 is derived by leaching of radiocarbon-dead carbon from deep mantle/crustal system.