

## Deep methane and helium emissions at convergent plate boundaries

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Methane plays an important role in the greenhouse effect of the atmosphere and in the chemistry of ozone reduction. Major methane emissions into the atmosphere originate from the biosphere (e.g., wetlands, rice paddies and animals), the geosphere (e.g., hydrocarbon basins and geothermal areas) and anthropogenic activity (e.g., natural gas production and distribution, coal mining). Natural methane emission from the geosphere is generally characterized by a radiocarbon-free signature (age > 50 kyrs) and may preserve information on the deep fluid in the Earth's crust. In this work, the origin of methane in forearc and volcanic arc regions at subduction zones, and collision zone at convergent plate boundaries is discussed using the carbon isotope composition of methane together with the abundances and isotope signatures of associated volatile elements such as helium, argon and nitrogen. We collected methane-rich natural gas samples in the South Kanto gas field in Japan and from mud volcanoes in South Taiwan. For comparison, we acquired natural gases in the Akita and Niigata gas fields located in the volcanic arc region of Japan. Chemical composition ( $\text{CH}_4$ ,  $\text{C}_2\text{H}_6$ ,  $\text{C}_3\text{H}_8$ ,  $\text{CO}_2$ ,  $\text{N}_2$ ,  $\text{O}_2$ , Ar, and He abundances), carbon and nitrogen isotope signatures ( $^{13}\text{C}/^{12}\text{C}$  of  $\text{CH}_4$ ,  $^{15}\text{N}/^{14}\text{N}$ ), noble-gas isotope ratios ( $^3\text{He}/^4\text{He}$ ,  $^4\text{He}/^{20}\text{Ne}$ , and  $^{40}\text{Ar}/^{36}\text{Ar}$ ) were measured using a quadrupole mass spectrometer, a continuous flow GC-IRMS system, and a noble gas mass spectrometer, respectively. The methane-rich gas in the South Kanto gas field shows a typical microbial signature characterized by light carbon isotopes, high  $\text{CH}_4/(\text{C}_2\text{H}_6 + \text{C}_3\text{H}_8)$  and low  $^3\text{He}/^4\text{He}$  ratios, while the natural gases in the Akita and Niigata region show a thermogenic signature with prevalence of heavy carbon isotopes, low  $\text{CH}_4/(\text{C}_2\text{H}_6 + \text{C}_3\text{H}_8)$  and high  $^3\text{He}/^4\text{He}$  ratios. These observations are consistent with those reported in the literature. On the other hand, methane-rich gases from mud volcanoes in South Taiwan show heavy carbon isotopes,  $\text{CH}_4/(\text{C}_2\text{H}_6 + \text{C}_3\text{H}_8)$  ratios between microbial and thermogenic signatures and variable  $^3\text{He}/^4\text{He}$  ratios, a part of which cannot be explained by a simple binary mixing of microbial and thermogenic methane. We also measured helium and carbon isotopes of submarine hydrothermal systems close to the Tokara Islands and within Kagoshima Bay. Gas geochemistry of the collected seawater and porewater samples is compared with those from the Japan Trench and the Nankai Trough. Finally, all methane-rich gases from submarine and on-land expressions of fluid emission are discussed within the frame work of the respective geotectonic settings.

Keywords: methane gas, helium isotope, plate boundary