Adsorbed methane in a Miocene methane-seep carbonate from Joetsu, Japan

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Submarine cold seeps are major sources of methane, a main component of natural gases and a potent greenhouse gas. Methane is generated by microbial or thermal degradation of organic matter in the shallow or deep subsurface, respectively, and the origins of methane can be determined through geochemical analyses including stable carbon isotopes. Identifying the origin of methane in ancient seep fluids should provide an insight into the origins and pathways of seep fluids in the geologic past, but a direct way to estimate this has not yet been established. In modern and ancient methane seeps, anaerobic oxidation of methane (AOM) by microbial consortia increases alkalinity and induces methane-derived authigenic carbonate precipitation. In this study, we extracted "adsorbed gas" [1] by acid dissolution of ancient methane-seep carbonate samples collected from the upper Miocene Nodani Formation in Joetsu City, Niigata Prefecture, central Japan. We then analysed the stable carbon isotopic compositions ($\delta^{13}C$ values) of the extracted gas to estimate its origin. As a result, we successfully extracted methane from all samples and found $\delta^{13}C$ values ranging from −60.7 to −40.0‰. This suggests that the extracted methane is composed of a mixture of biogenic and thermogenic methane in various ratios and/or contains biogenic methane enriched in ^{13}C due to fractionation through AOM. A weak but significant positive correlation was found in the $\delta^{13}C$ values between the extracted methane and its host carbonate cements. This correlation implies that the methane contained in the ancient seep fluid was trapped in the host carbonate cements during their formation. Because it is also possible that the methane was adsorbed after precipitation and during burial of the carbonate, further investigation of the preservation state of the gases in the seep carbonate is required to determine the origin of methane in the ancient seep fluid.

[1] Ijiri et al. (2009) *Geo-Mar Lett* **29**, 301–308.